Designer atoms : Engineering Rydberg atoms using pulsed electric fields

J. J. Mestayer, W. Zhao, J. C. Lancaster, F. B. Dunning

Rice University

C. O. Reinhold

Oak Ridge National Laboratory and University of Tennessee

S. Yoshida, J. Burgdörfer

Vienna Technical University

Rydberg atoms

- one electron excited to a state of large principal quantum number n
- physically very large - Bohr radius scales as n^2
- weakly bound - binding energy decreases as $1/n^2$

High-n atoms provide a mesoscopic quantum entity that bridges quantum and classical worlds

Motivation

- explore classical limit of quantum mechanics
- evaluate protocols for controlling and manipulating atomic wavefunctions
- examine concepts for quantum information processing in mesoscopic systems
- examine dephasing and decoherence
- gain insights into physics in the ultra-fast ultra-intense regime
- generate non-dispersive wavepackets

Engineer high *n* atoms using pulsed electric fields

Engineering Rydberg Wavepackets

Use pulsed unidirectional electric fields, termed half-cycle pulses (HCPs), of duration $T_p << T_n$

Each HCP delivers an impulsive momentum transfer or ”kick” to the electron

$$\Delta\vec{p} = -\int \vec{F}_{HCP}(t)dt$$

Create desired final state using tailored sequence of HCPs

Realization of Impulsive Regime

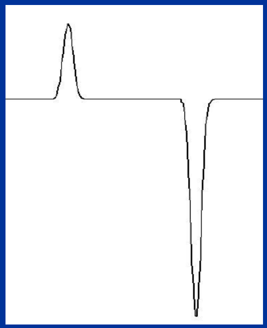
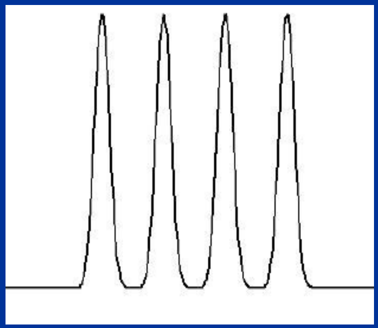
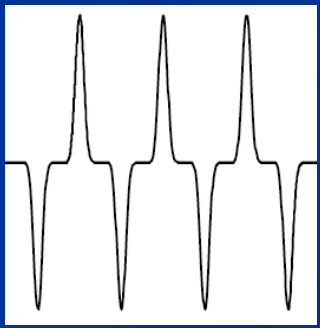
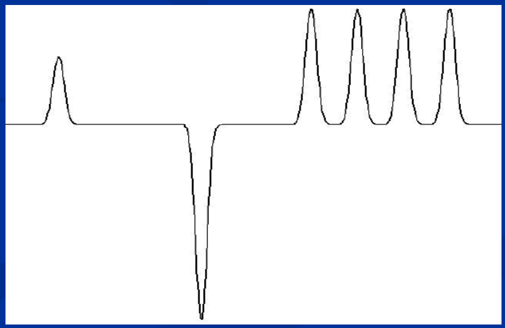
Electron orbital period $T_n = n^3 t_1$, where $t_1 = 1.5\times10^{-16}$ s

At $n = 30$, $T_n = 4\times10^{-12}$ s; $n = 300$, $T_n = 4\times10^{-9}$ s

Two approaches:

- use ultra-short ($T_p < 1$ps) freely-propagating HCPs generated by fs-laser-triggered photoconducting switch (Bucksbaum, Jones, Noordam, Stroud)
- use longer HCPs ($T_p > 500$ps) produced by applying output of pulse generator to a nearby electrode

easy to control and measure HCPs, and generate complex HCP trains

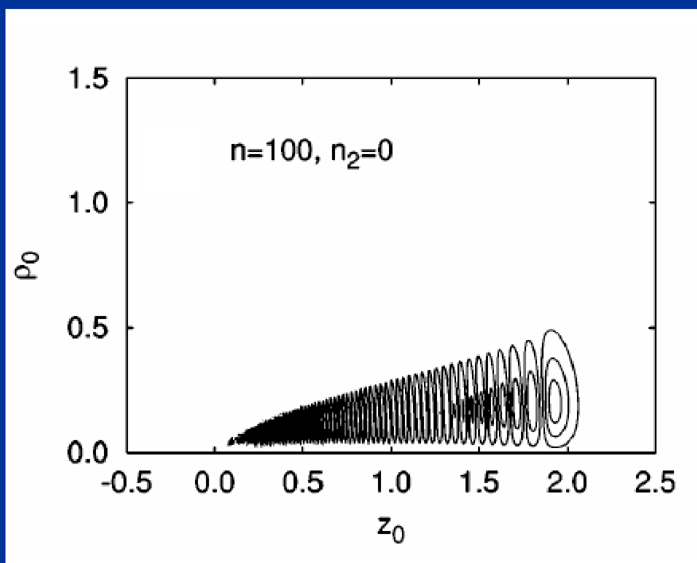





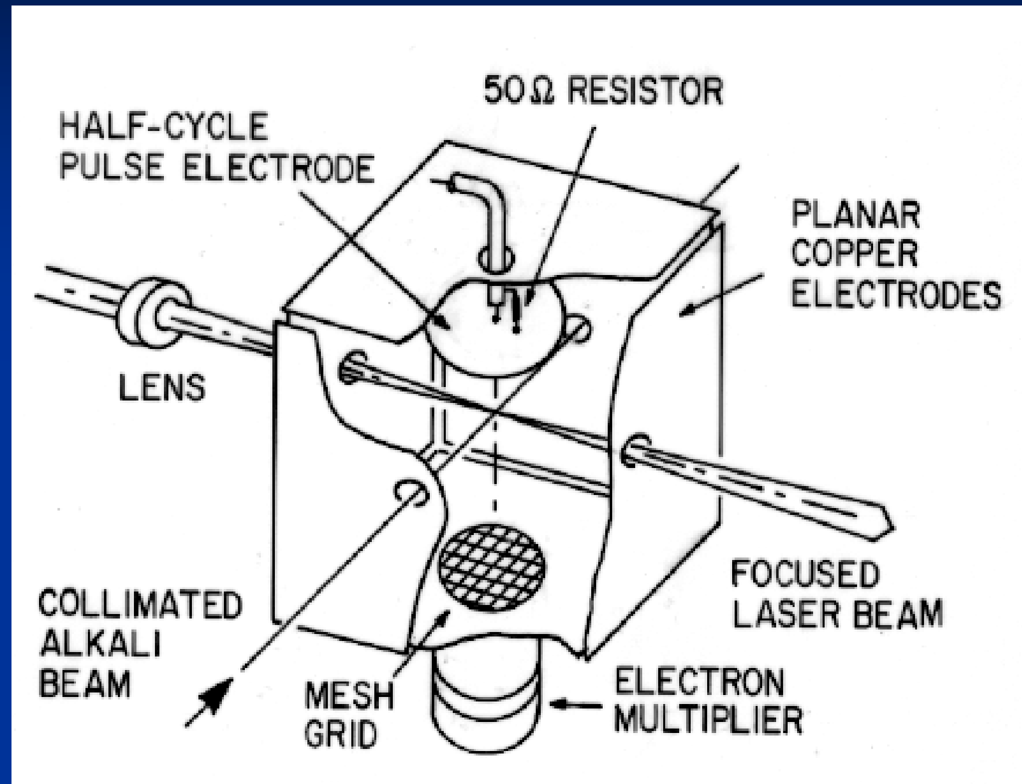
Need to work with very-high-n atoms, n > 350

Studies at very high *n*, *n* > 350

Difficult: Rydberg levels closely spaced, atoms strongly perturbed by external fields.

Produce quasi-1D atoms by exciting selected Stark states.





Effect of single HCP: $T_p << T_n$

Classically: impulse $\Delta\vec{p}$ changes electron energy by

$$\Delta E = \Delta E_k = \left(\frac{\vec{p}_i + \Delta\vec{p}}{2}\right)^2 - \frac{\vec{p}_i^{\,2}}{2} = \frac{\Delta p^2}{2} + \vec{p}_i \cdot \Delta\vec{p} = \frac{\Delta p_z^2}{2} + p_{iz}\Delta p_z$$

Leads to distribution of final n states or, if ΔE sufficient, to ionization.

Measurements of survival probability used to:

- monitor time evolution of p_{iz}
- map distribution of initial z-components of electron momentum

Quantum mechanically: impulse(s) produces coherent superposition of states, i. e., a wavepacket

$$\left|\Psi(t)\right\rangle = \sum_n e^{-iE_n t} \sum_\ell \left\langle n\ell m \middle| \Psi(0)\right\rangle \left|n\ell m\right\rangle \qquad \Psi(0) = e^{i\Delta\vec{p}\cdot\vec{r}}\left|\phi_i\right\rangle$$

Explore behavior of wavepackets using CTMC simulations

Wavepacket simulations

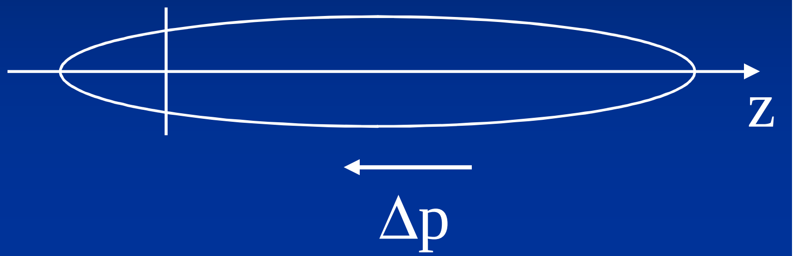
Employ classical-trajectory Monte Carlo (CTMC) method

- initial state represented by appropriate distribution of phase points
- track evolution of each phase point during HCP sequence by solving Hamilton's equation of motion

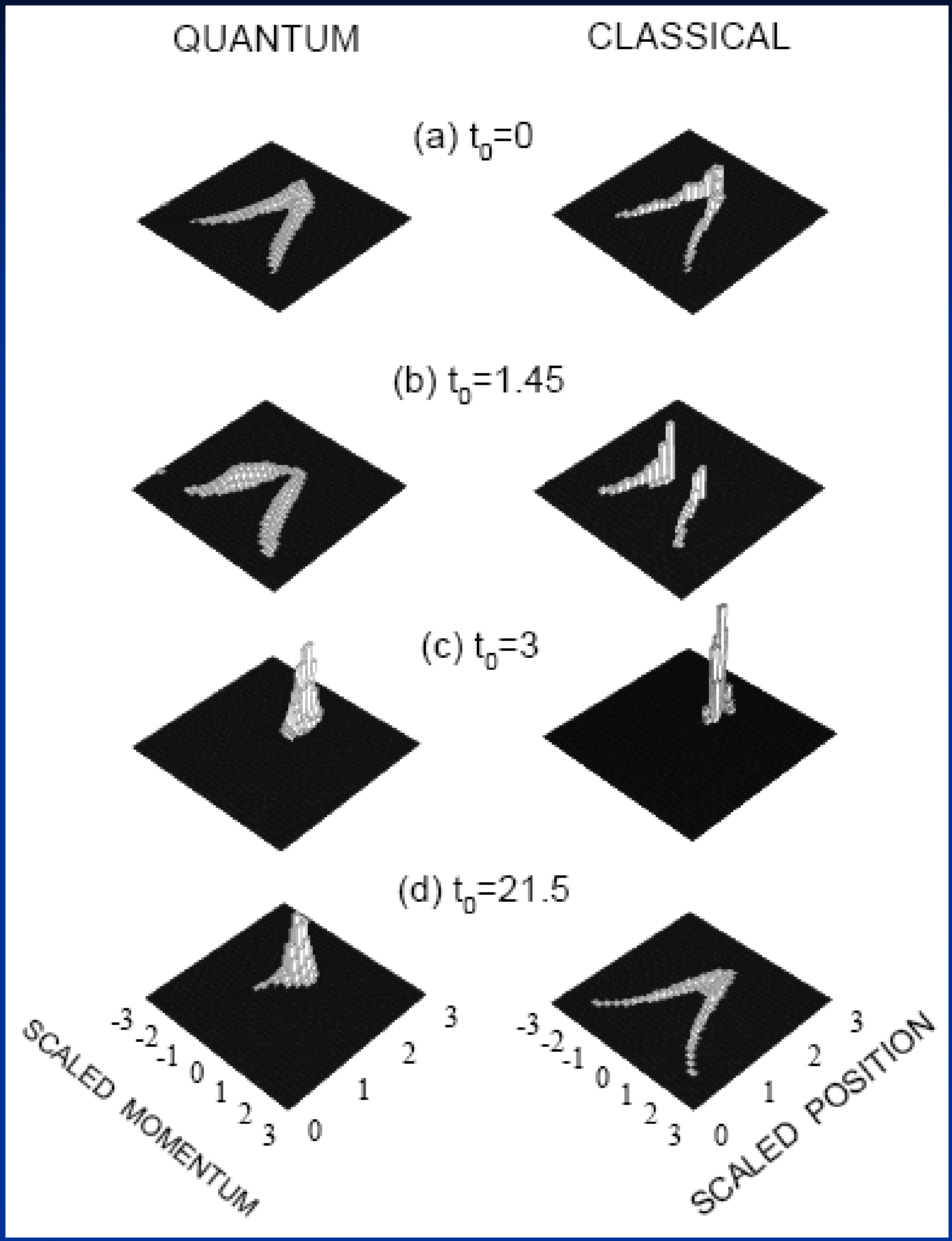
$$H(t) = \frac{p^2}{2} - \frac{1}{r} + zF_{train}(t)$$

- build up distribution of phase points at time of interest - mirrors probability density distribution of corresponding wavepacket
- consider different times to examine evolution of wavepacket

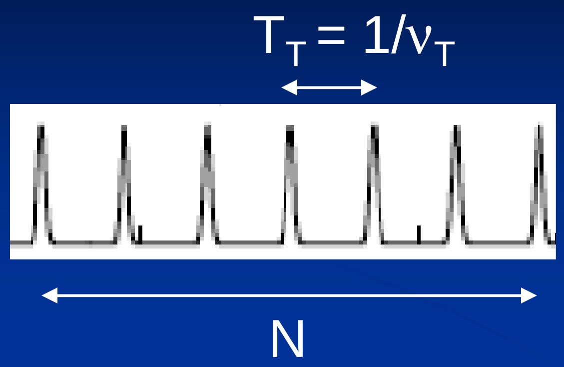
1D atoms - effect of a single HCP



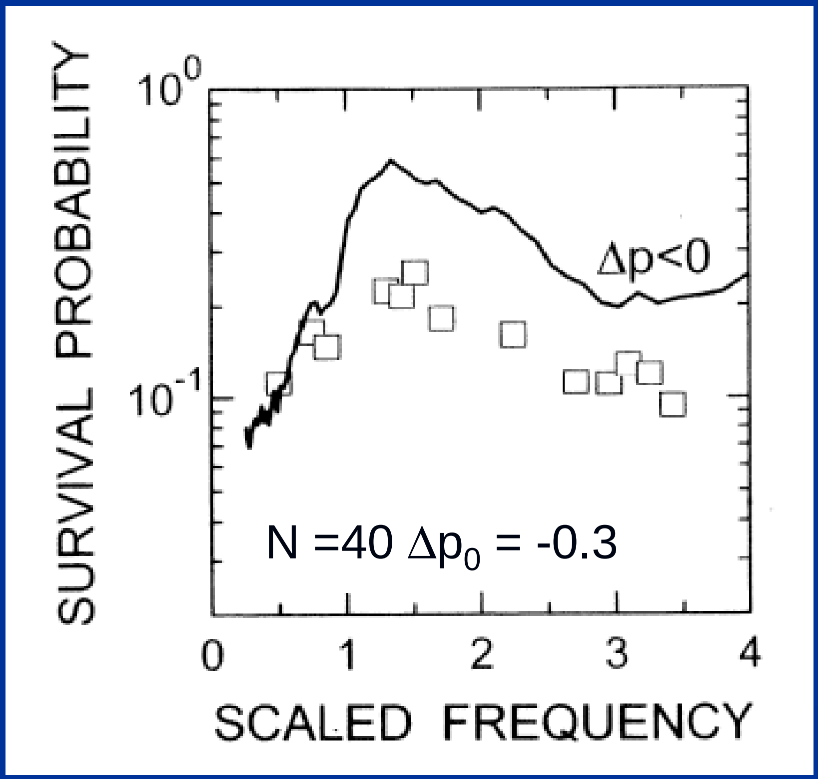
- induce strong transient phase-space localization
- observed with quasi-1D atoms
- great starting point for further manipulation



1D atoms - effect of multiple periodic HCPs



Impulses all applied in same direction -might expect series of energy transfers leading to ionization



- large fraction of atoms survive
- peak in survival probability seen at $\nu_T \sim 1.3\ \nu_n$

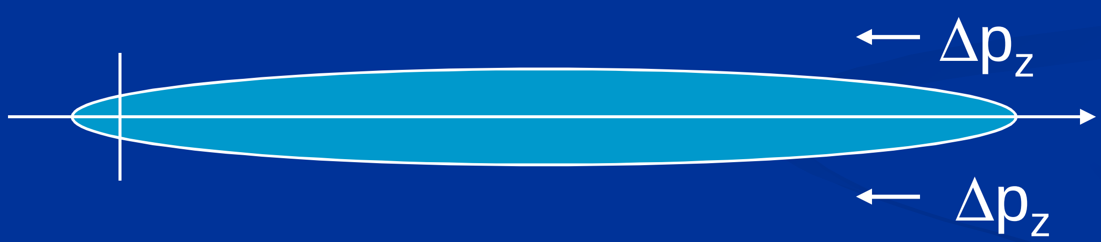
Origin of stabilization?

Dynamical stabilization

To survive many HCPs, each must transfer little energy to electron, i.e., require:

$$\Delta E = \Delta p_z^2/2 + p_{iz}\Delta p_z = 0 \quad \Rightarrow \quad p_{iz} = -\Delta p_z/2, \quad p_{fz} = +\Delta p_z/2$$

p_z must then evolve through orbital motion to $-\Delta p_z/2$ at time of next HCP



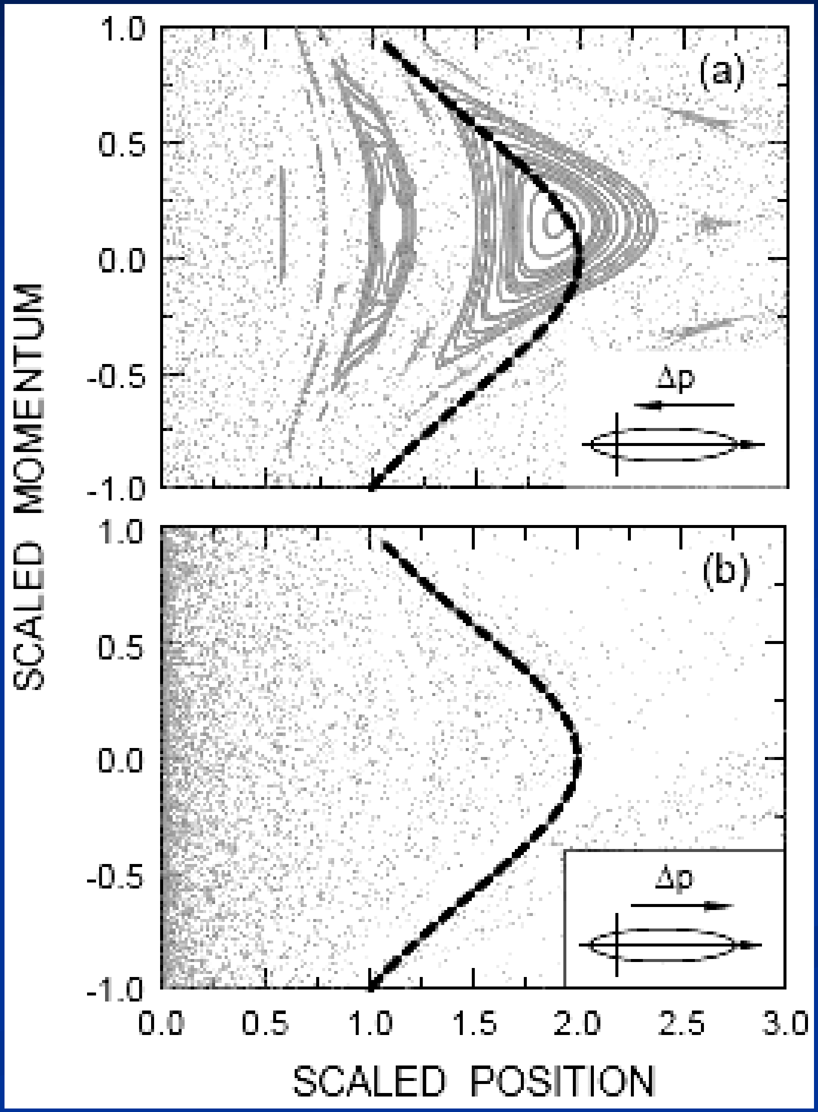
If electron motion synchronized with HCP frequency obtain dynamical stabilization - see by considering phase space for kicked atom

Phase space for periodically-kicked 1D atom

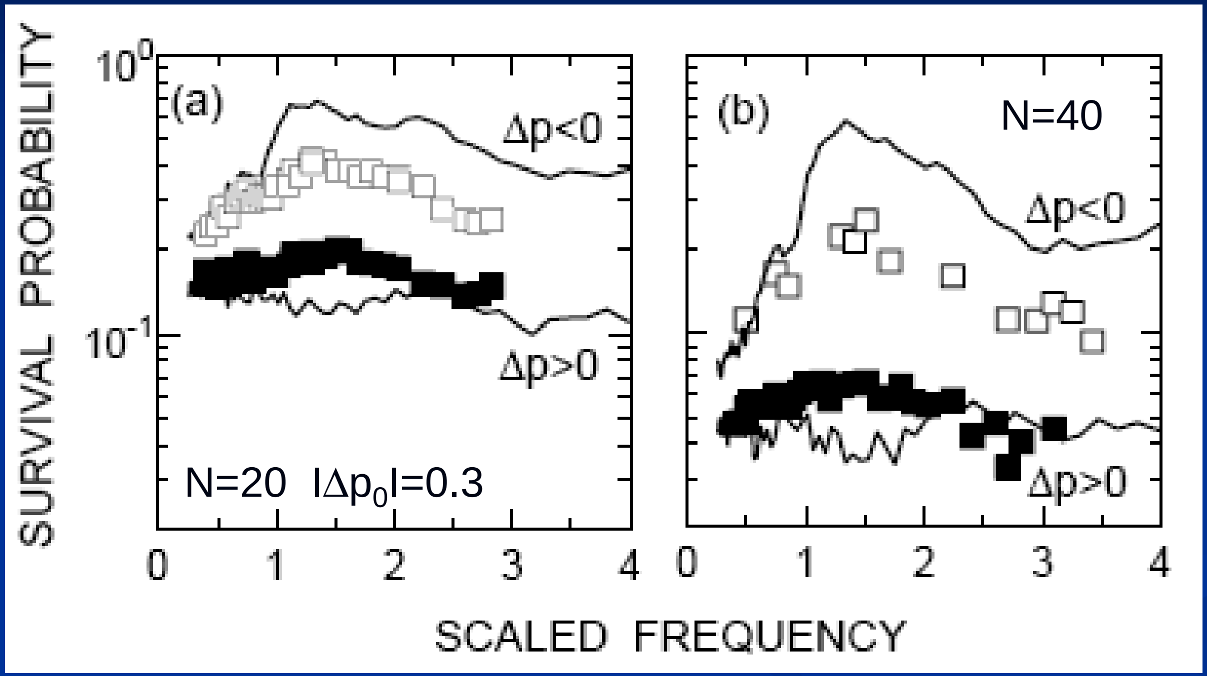
Poincare surfaces of section

- for $\Delta p < 0$ see islands of stability embedded in chaotic sea
- for $\Delta p > 0$ system globally chaotic
- if initial phase point lies in island remains trapped and survives large number of kicks
- produce non-dispersive wavepacket that undergoes transient phase space localization

Strong asymmetry confirmed by experiment



Effect of multiple HCPs: Quasi-1D *n* = 350 atoms

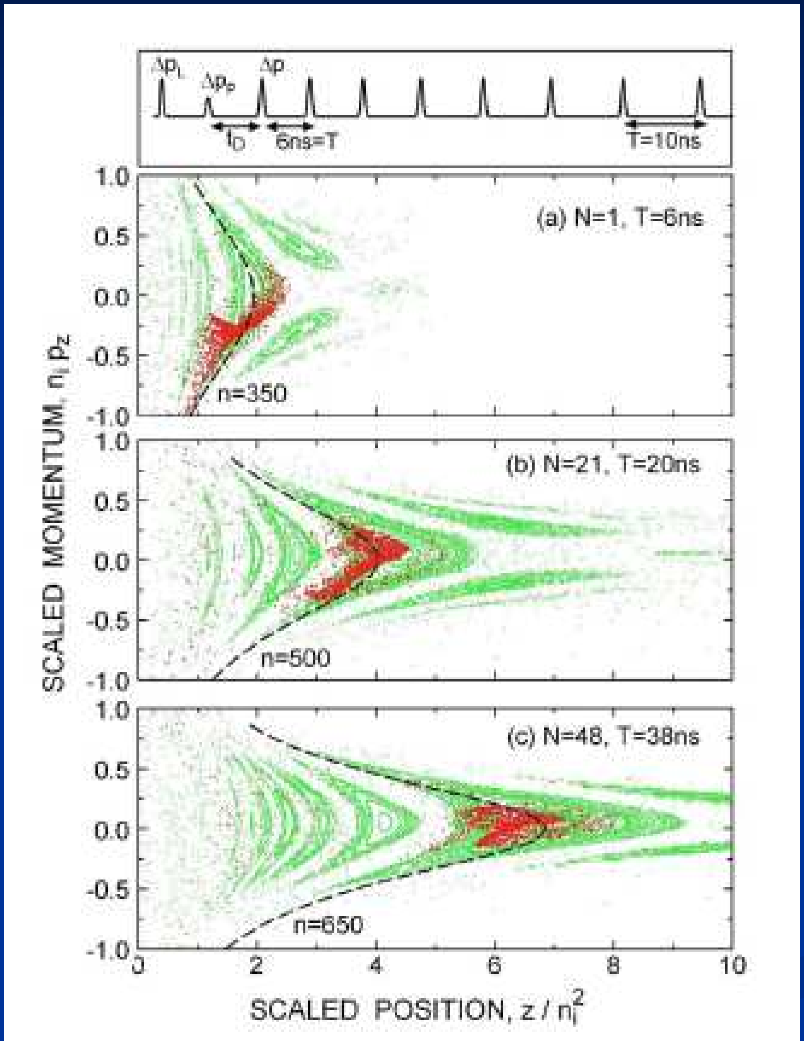


- pronounced asymmetry in survival probability
- survival probability large - wavepacket trapped for extended periods
- trapping provides opportunity for navigating in phase space

Navigating in phase space

Position of islands depends on kick size and frequency

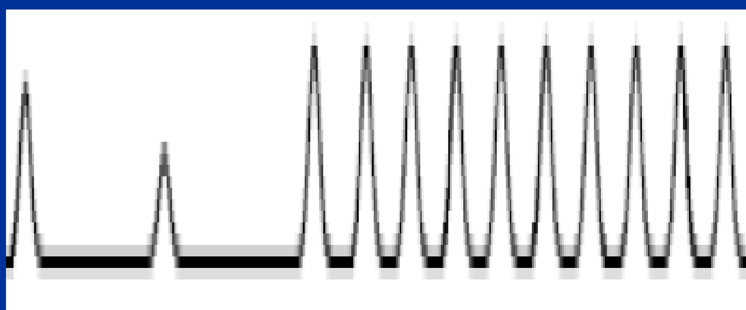
- steer island away from nucleus by “down chirping” kick frequency

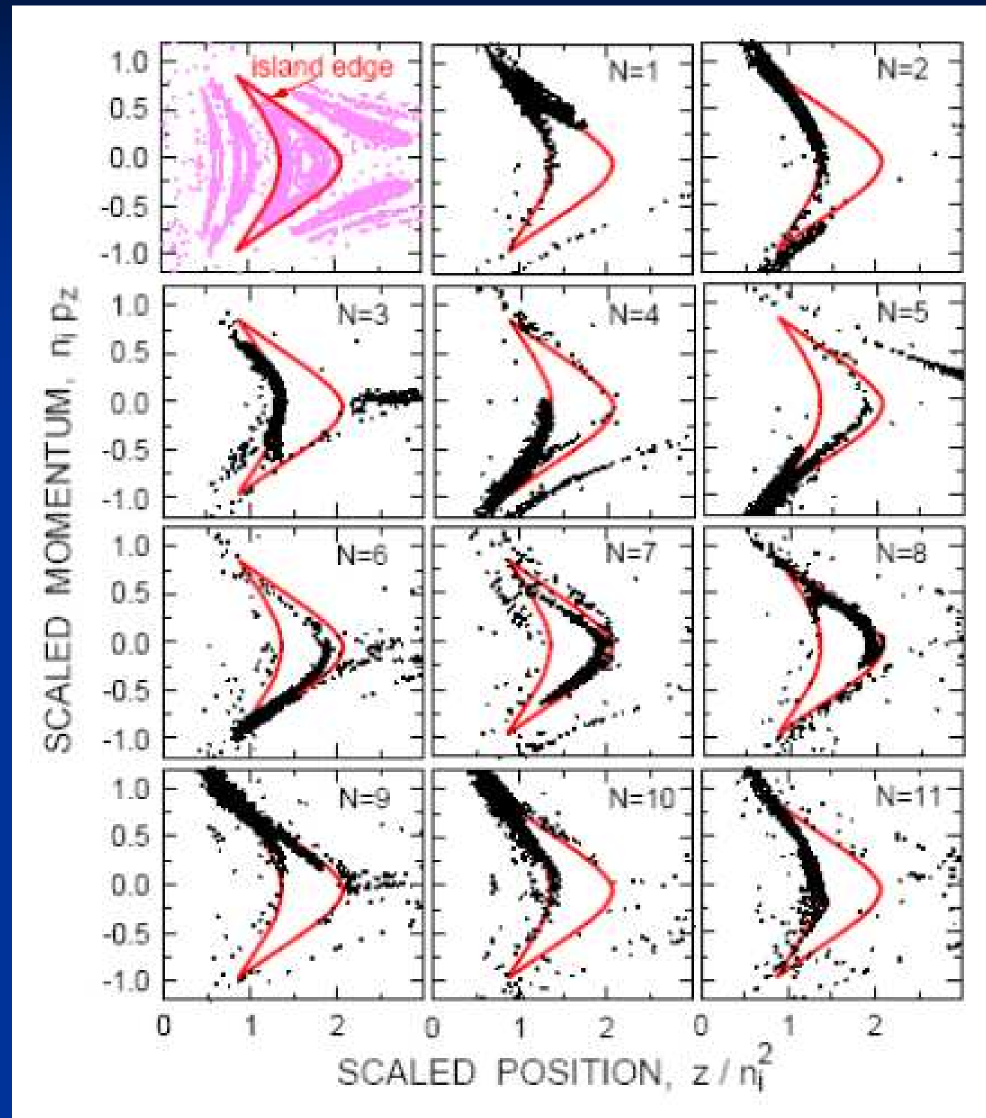


Can control atomic wavepackets using periodic HCP trains - key lies in initial island loading

Selective island loading: CTMC simulations

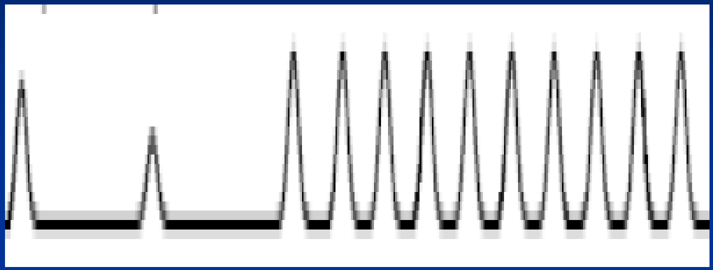
Take transiently localized state - place at center or periphery of largest island by varying island position , i e, T_T, and t_d

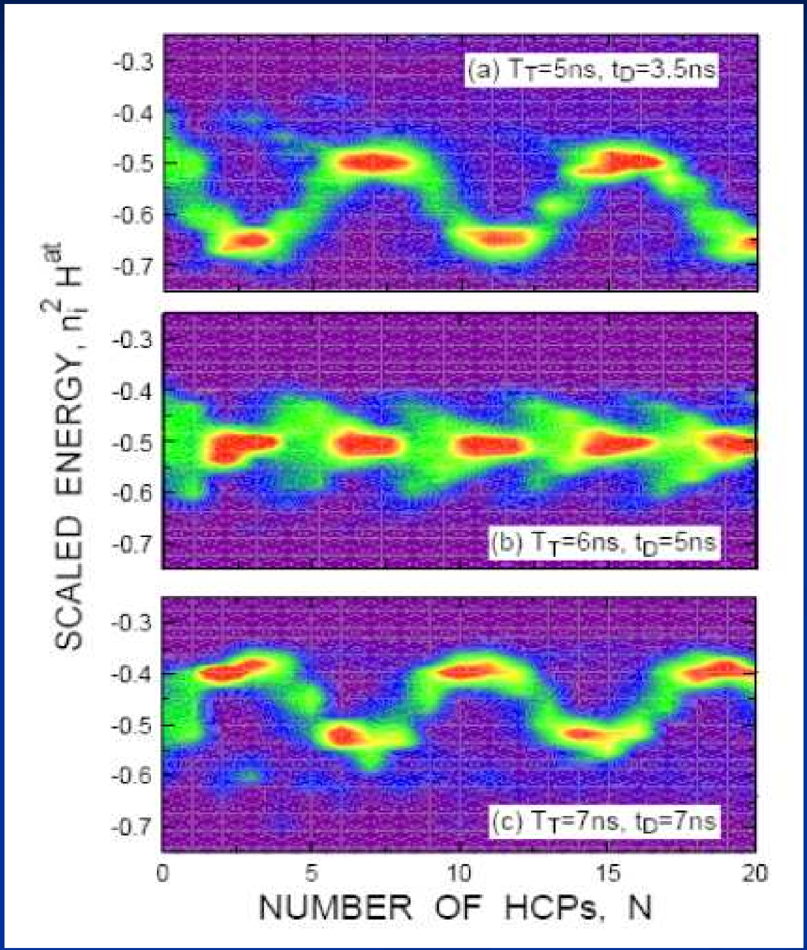




- wavepacket circumnavigates island as N increases
- leads to periodic changes in electron energy

Selective island loading: electron energy distribution



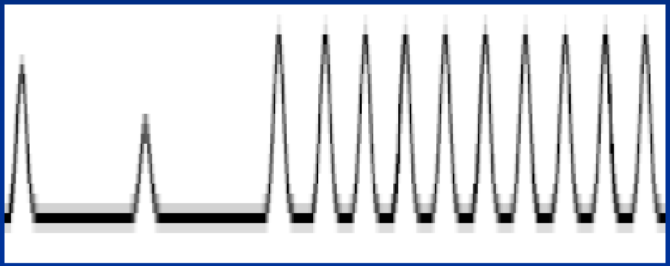


- motion around periphery gives periodic variations in energy
- persist to high N
- fluctuations minimal if load center

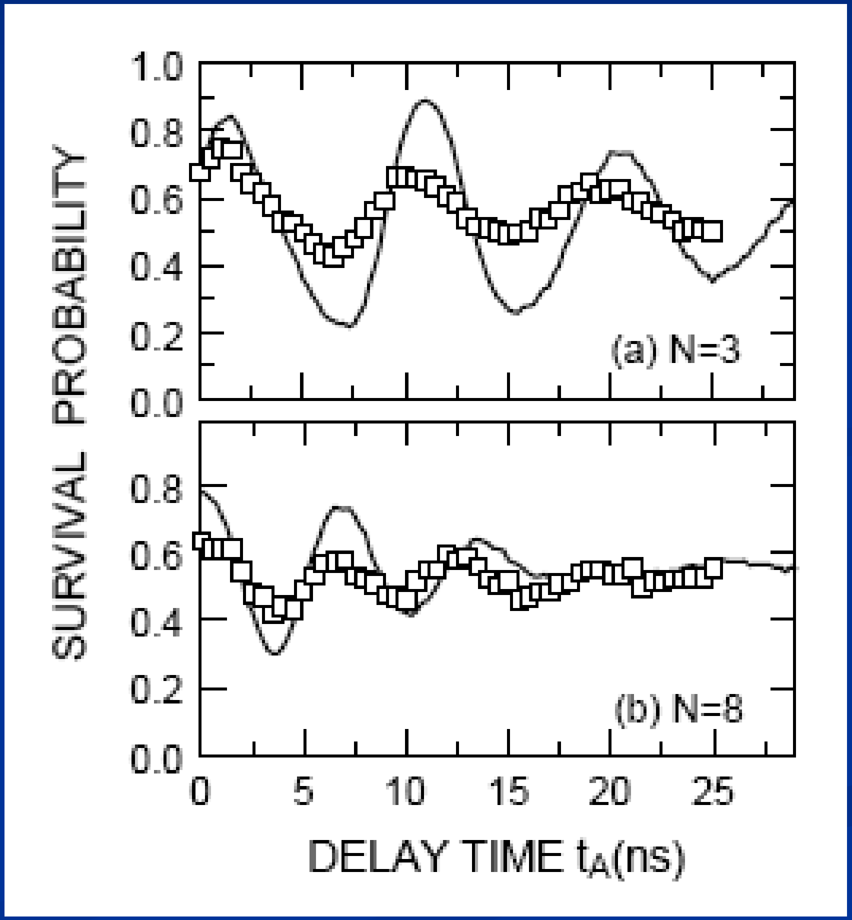
Observe changes in final energy (or *n*) distribution with N using a probe pulse

Selective island loading: final *n* distribution evolution

T_T = 7ns, t_D = 7ns



- time evolution characteristic of final *n* distribution
- period varies from ~9.5ns after N=3 HCPs to ~6ns after N=8 HCPs
- CTMC simulations in accord with experiment

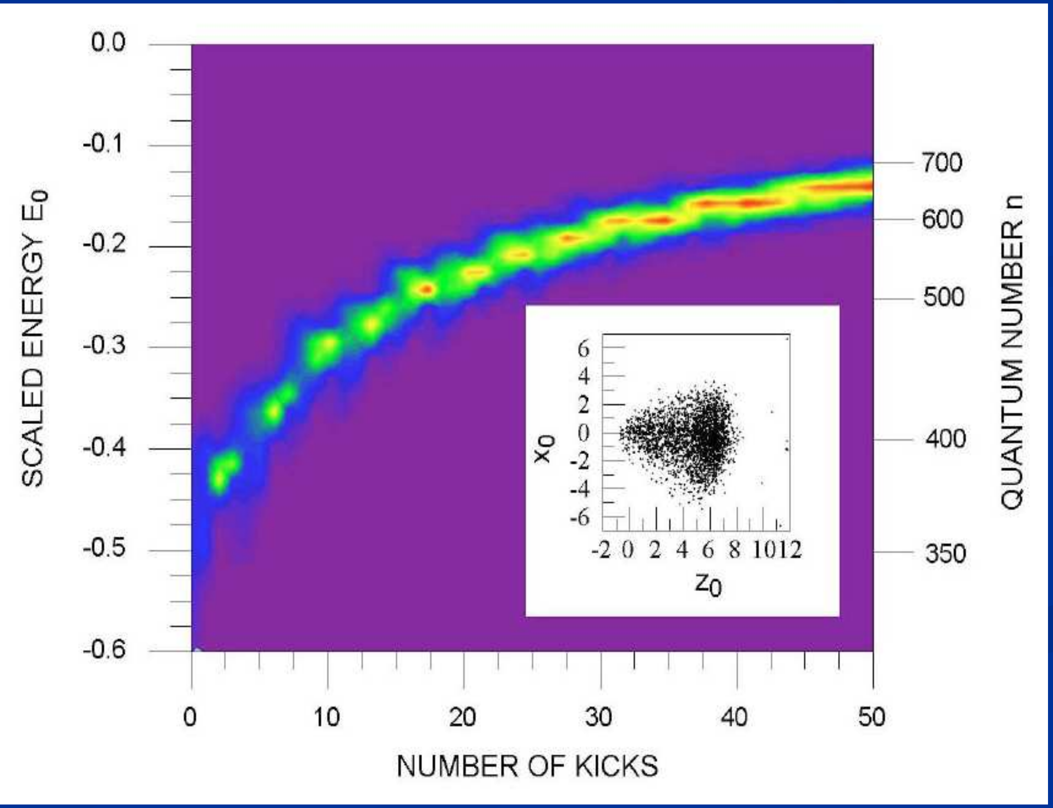


Navigating in phase space: Chirped HCP Train

- load phase-space-localized n = 350 wavepacket into stable island
- down chirp HCP frequency to drive to targeted final n state

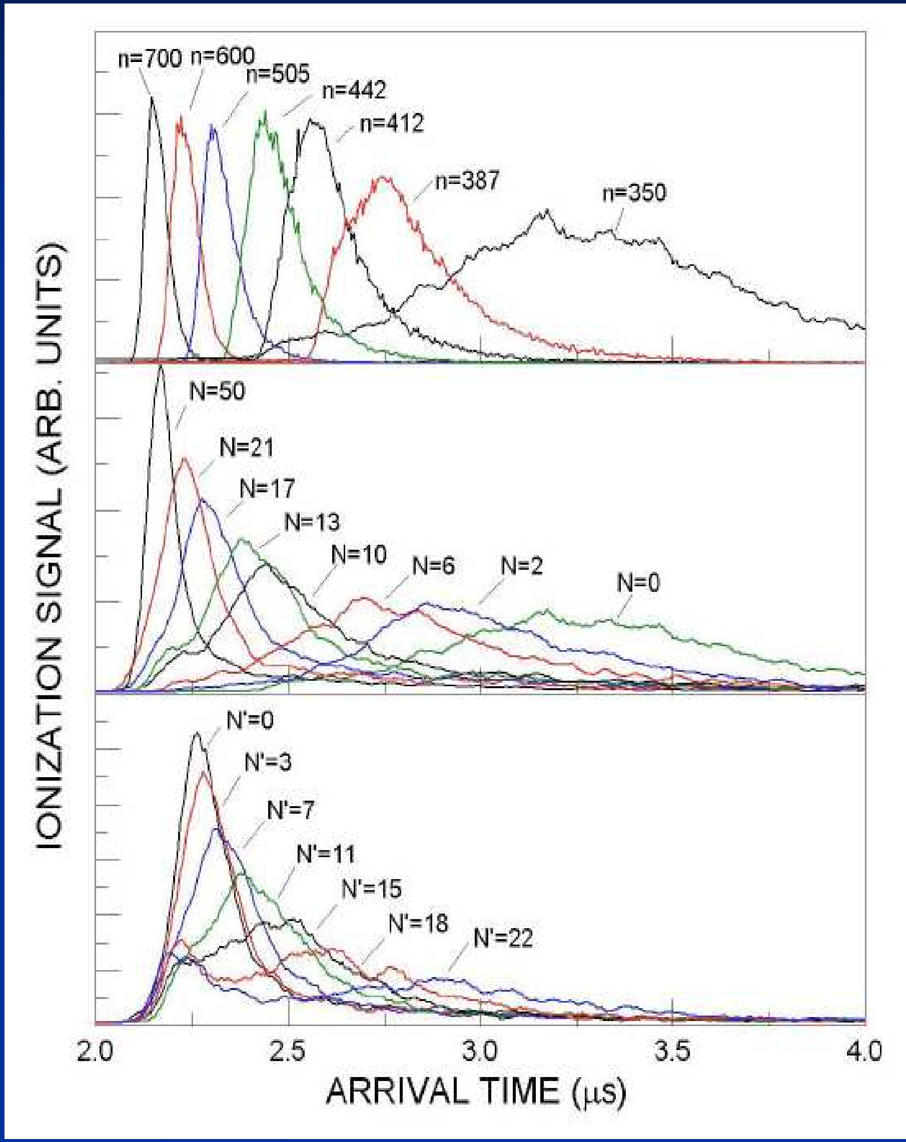
ΔT (N - N+1) =
5.33 + 0.67N ns

- wavepacket remains trapped
- narrow final n range
- final state strongly polarized



Evolution of *n* distribution

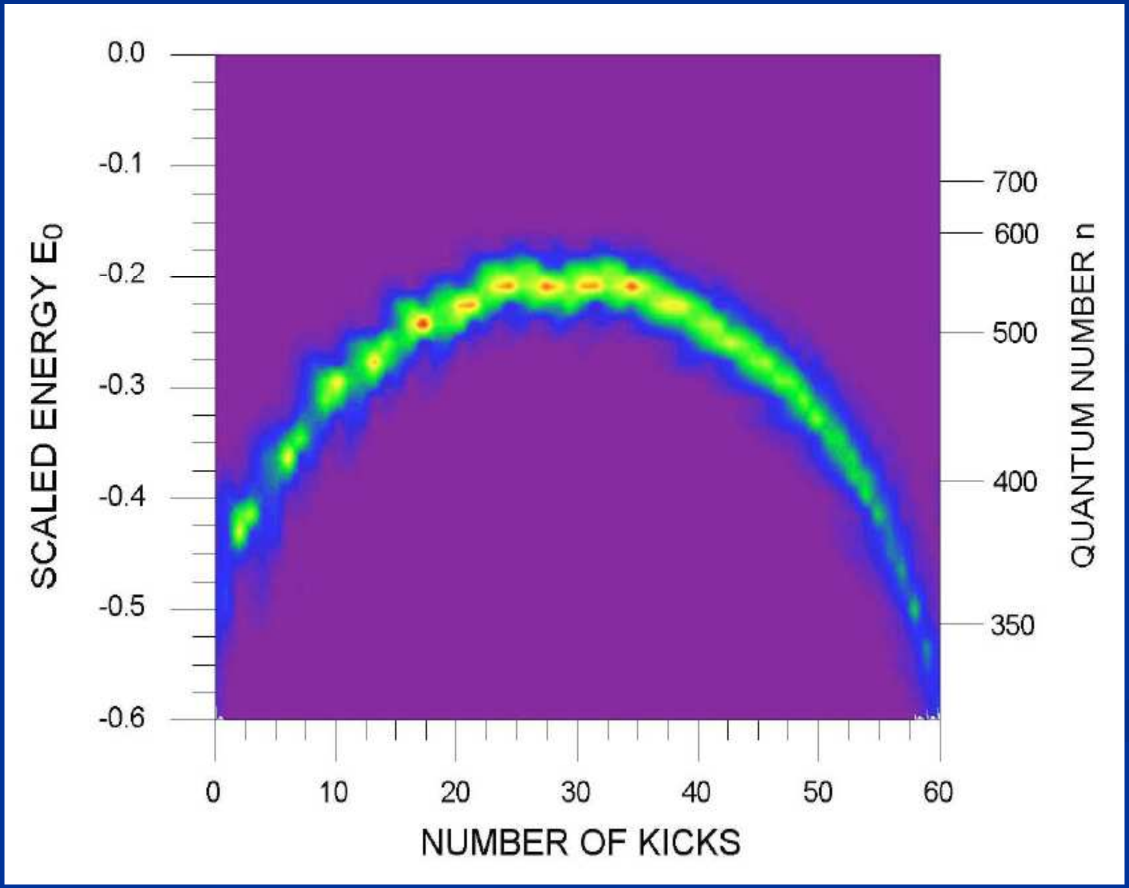
- monitor using SFI
- as N increase spectra move to higher *n*
- final *n* distribution narrow, $\Delta n \sim \pm 20$ centered at $n \sim 670$
- by reversing chirp can move to lower *n*



Demonstration of control

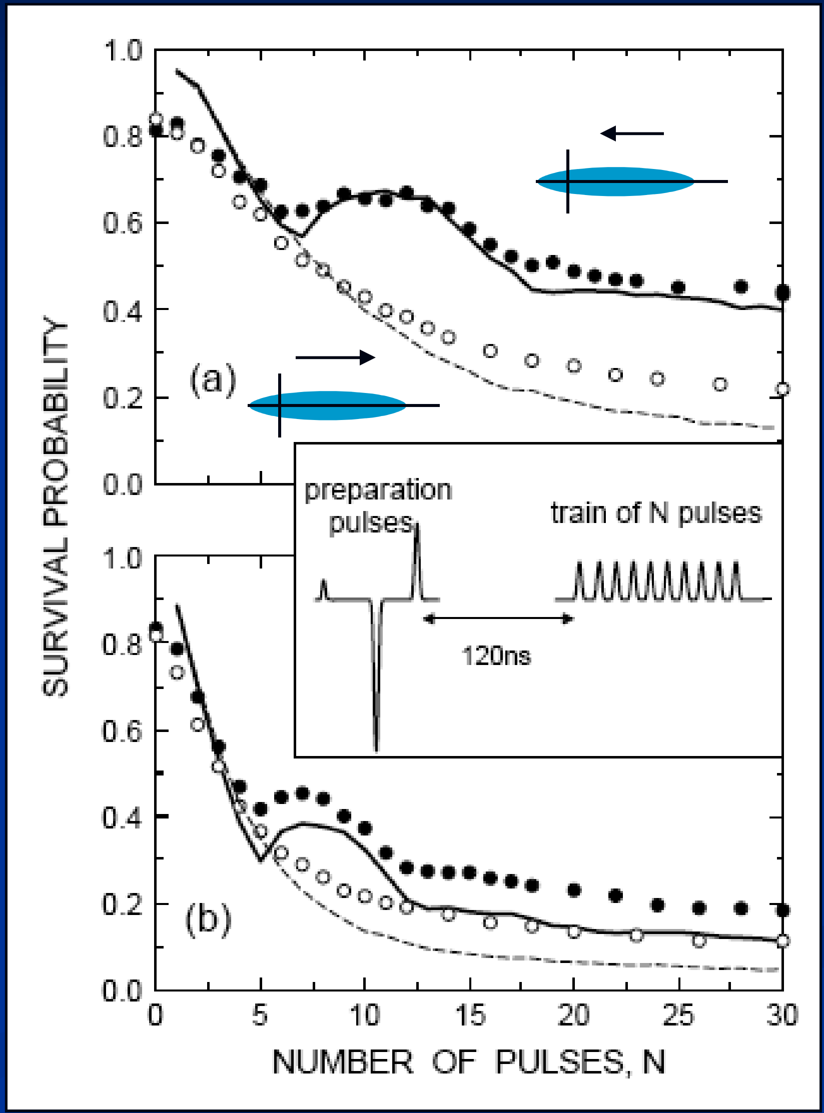
Linearly increase ΔT for 25 HCPs, hold constant for 10 HCPs, linearly decrease for 25 HCPs.

Engineer quasi-1D states of arbitrarily high *n*



High scaled frequencies: N dependent survival probabilities

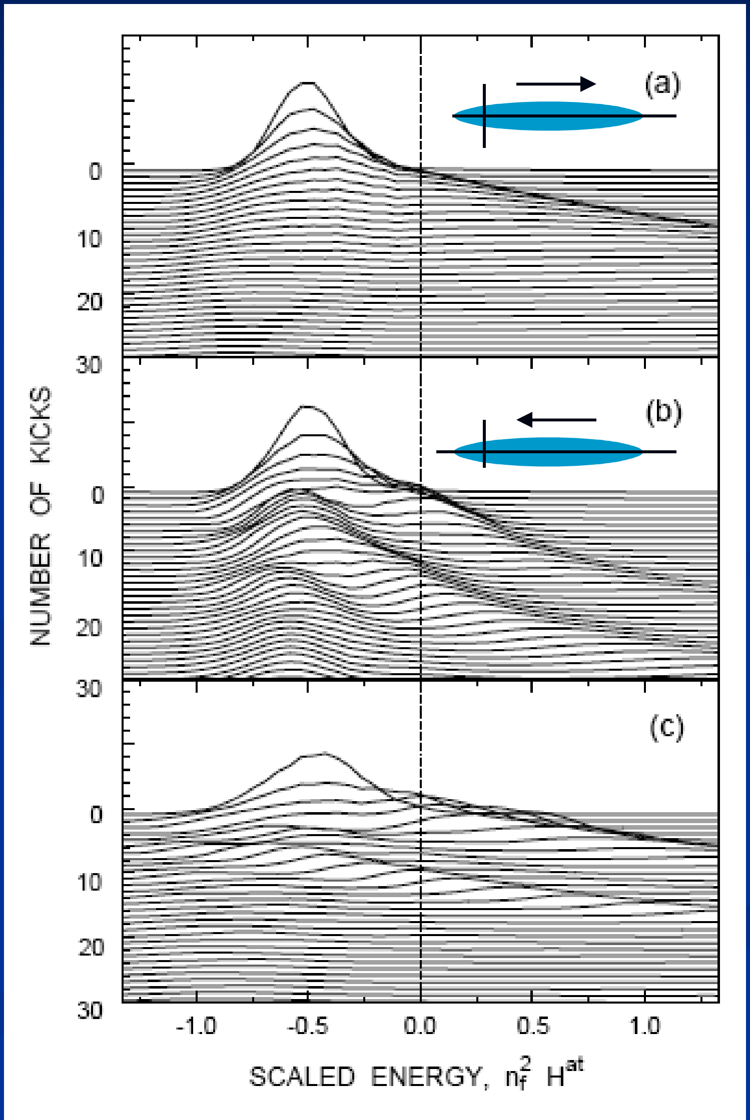
- behavior sensitive to kick direction
- pronounced non-monotonic structure in survival probability
- survival probability can increase with N



Behavior understood with aid of CTMC simulations

High scaled frequencies: energy distribution evolution

- for $\Delta p>0$ energy distribution broadens, moves to higher *n*
- for $\Delta p<0$ see series of “waves” passing into continuum
- features due to multiple scattering at core ion
- behavior parallels that in dc field

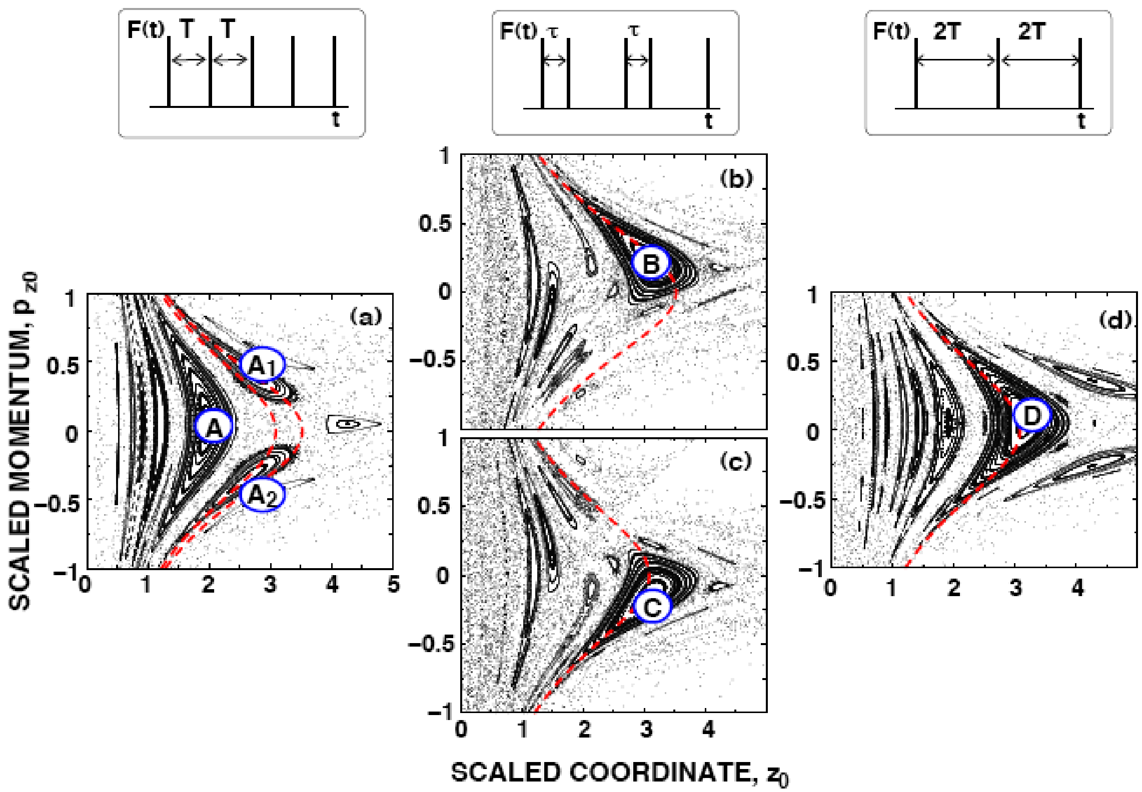


Transferring wavepackets between islands

Transfer from period-1 island A to period-2 islands A_1 and A_2

Protocol

- prepare wavepacket localized in A
- downchirp to populate D
- superpose second identical HCP train
- vary time delay τ to regenerate original HCP train

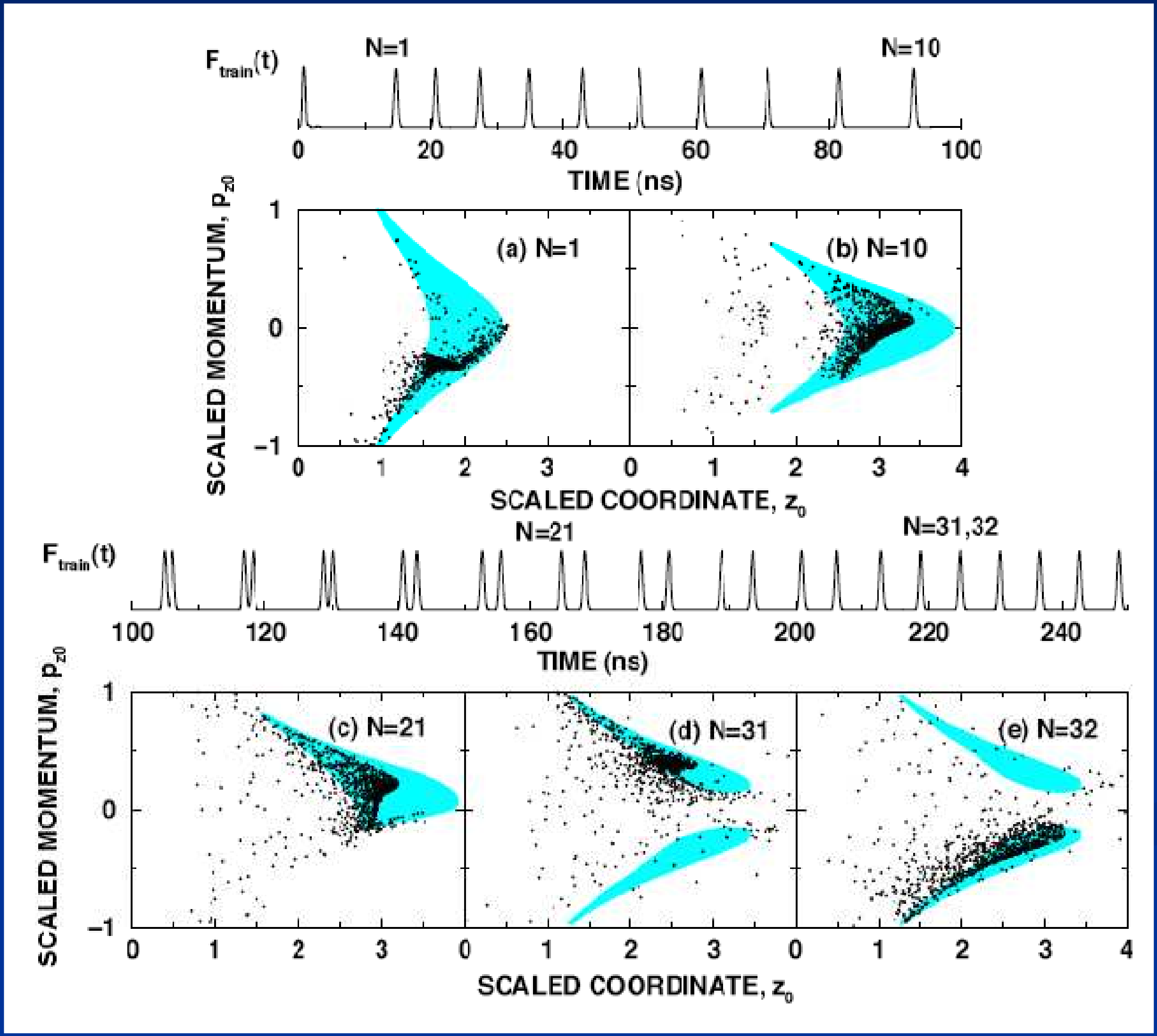


Use control variable τ to “morph” islands

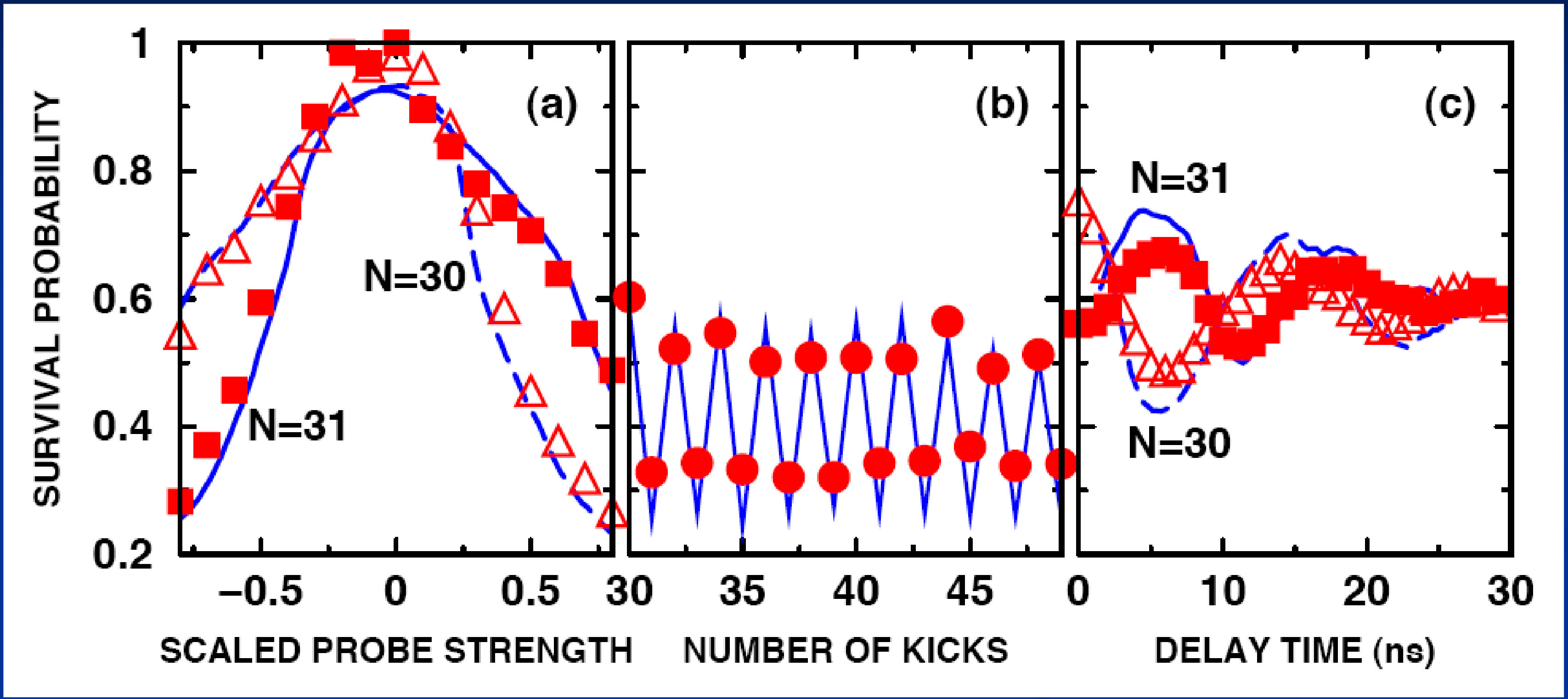
Wavepacket evolution - CTMC simulations

Consider maximally-polarized n = 350 Stark state

- initial wavepacket efficiently transferred to period-2 islands
- islands have different momenta - discriminate using probe HCP



Experimental results - quasi-1D *n* = 350 atoms



- sizable asymmetry in survival probability
- reverses with sign of probe kick
- survival probability oscillates with N for N > 31
- clear evidence of period-2 island population

Wavepacket dephasing

Two causes:

- wavepacket components evolve at different rates - dephases but remains fully coherent enabling revivals - coherent dephasing
- stochastic external perturbations like noise or collisions - leads to irreversible dephasing of wavepacket - decoherent dephasing or decoherence

Decoherence of fundamental importance for all potential carriers of quantum information

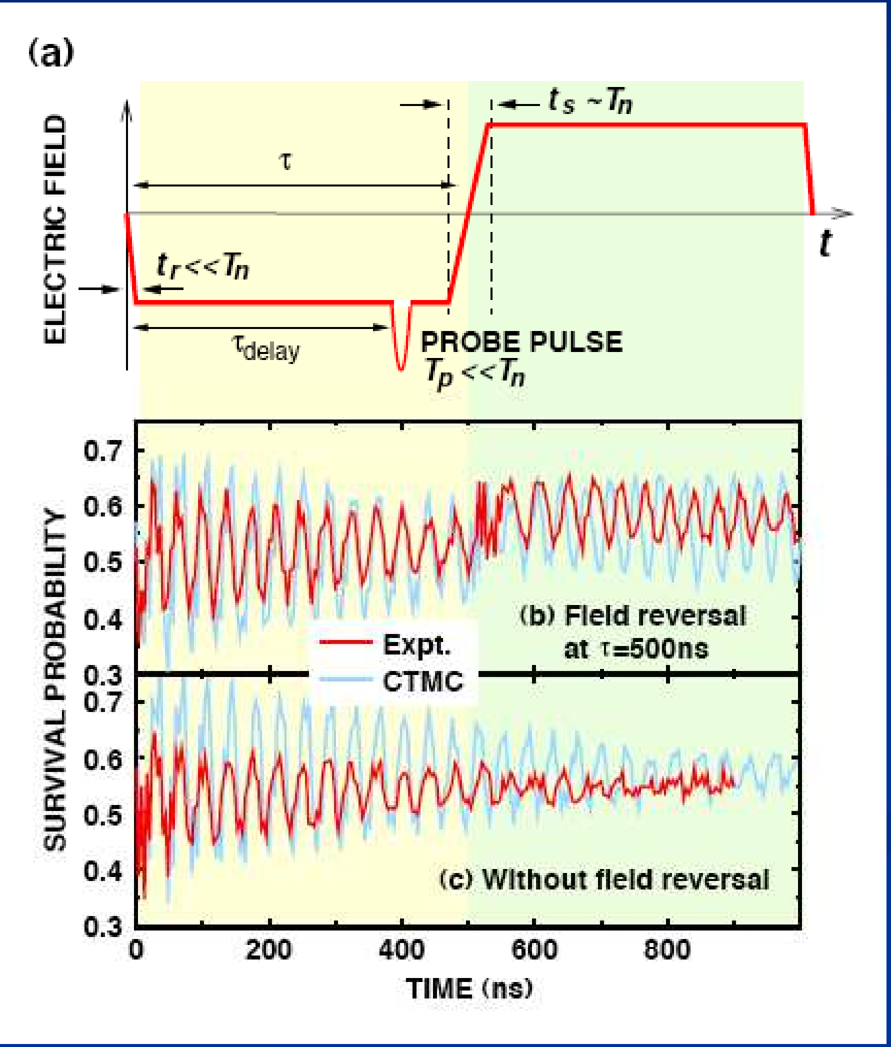
Study using a technique that involves electric dipole echoes in Stark wavepackets

Electric dipole echoes

Observe echoes in electric dipole moment of ensemble of Rydberg atoms precessing in an external field after its reversal - analogous to NMR

- produce quasi-1D atoms aligned along x axis
- apply pulsed dc field along z axis to create Stark wavepacket
- monitor wavepacket evolution with probe HCP - see series of quantum beats

If reverse field at t = τ observe strong quantum beat echo at t ~ 2τ - in accord with CTMC simulations

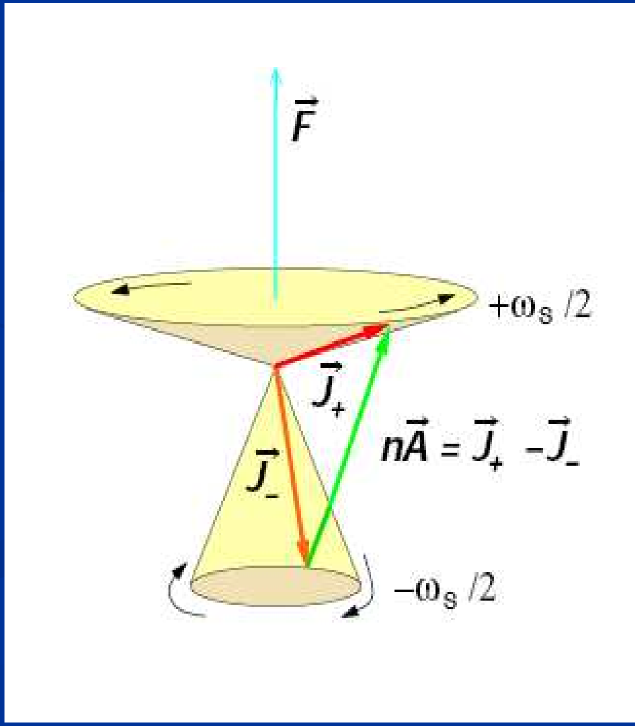


Evolution of Stark states

- classically, electron orbit characterized by energy E, angular momentum $\boldsymbol{L} = \boldsymbol{r} \times \boldsymbol{p}$, and Runge-Lenz vector $\boldsymbol{A} = \boldsymbol{p} \times \boldsymbol{L} - \boldsymbol{r}/r$
- in weak field F, ***L*** and ***A*** precess slowly - describe using orbit - averaged values <***L***>, <***A***>
- define two pseudo-spins $\boldsymbol{J}_{\pm} = 1/2\,(\langle \boldsymbol{L} \rangle \pm n \langle \boldsymbol{A} \rangle)$ - evolve according to effective Bloch equations

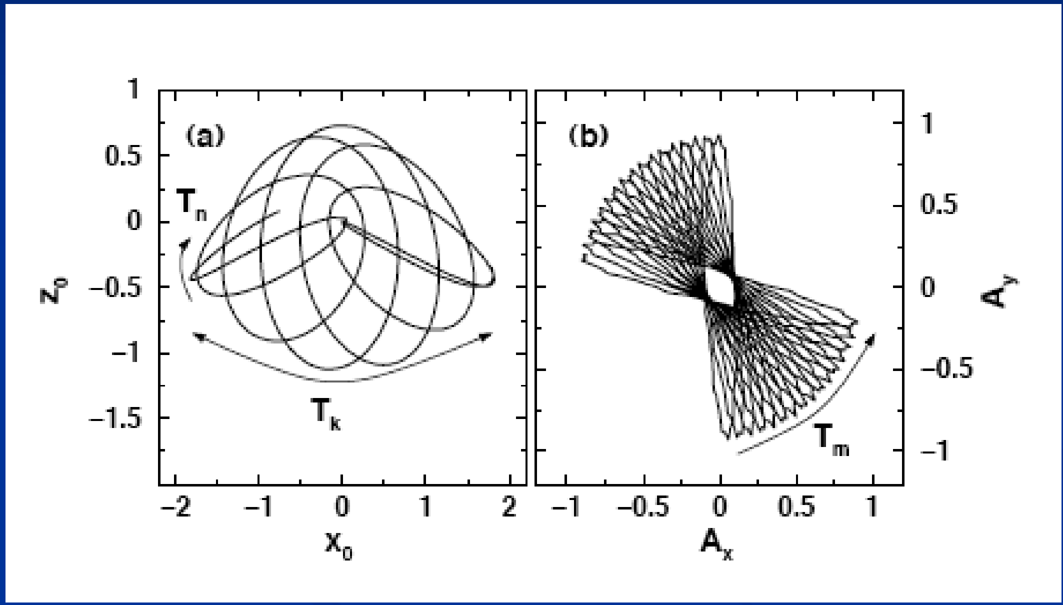
$$\frac{d}{dt}\vec{J}_{\pm} = \omega_{\pm}(F)\vec{J}_{\pm} \times \hat{z}$$

- $\boldsymbol{J}_+$, $\boldsymbol{J}_-$ precess in opposite directions about field
- magnitude of dipole moment varies periodically



Electron Motion in a Weak Field

Motion on three timescales:



- electron orbits rapidly on ~Kepler ellipse - period T_n
- ellipse precesses in field undergoing oscillations in eccentricity - period T_k
- plane of motion slowly rotates about z axis - period T_m . Shown by motion of Runge-Lenz vector

$$\boldsymbol{A} = \boldsymbol{p}\times\boldsymbol{L} - \boldsymbol{r} / r$$

Probe HCP maps variation of orientation and elongation of Kepler ellipse

Psuedo-spin Precession Frequencies

Hydrogenic energies

$$E_{n,k,m} = -\frac{1}{2n^2} + \frac{3}{2}nkF - \frac{1}{16}n^4\left[17n^2 - 3k^2 - 9m^2 + 19\right]F^2$$

Expressing classical energies in terms of $J_{\pm}^{z}$ = (m ± k)/2 obtain

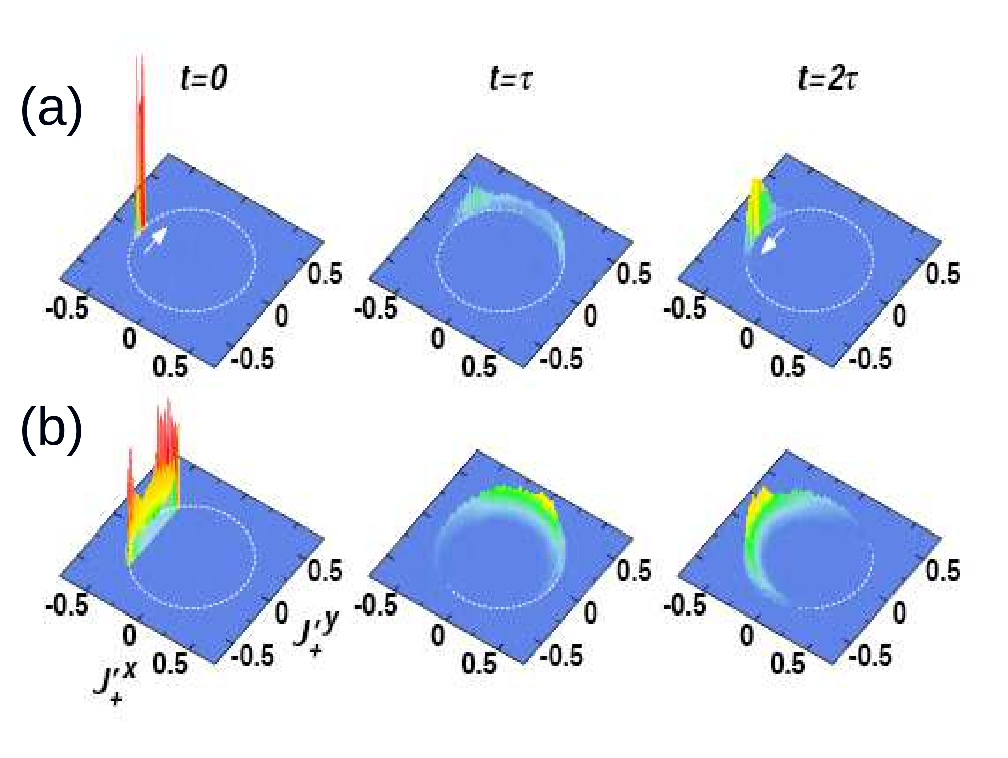
$$\omega_{\pm}(F) = \frac{\partial E(n, J_{+}^{z}, J_{-}^{z})}{\partial J_{\pm}^{z}} = \pm\left(\omega_k^{(1)}(F) + \omega_k^{(2)}(F)\right) + \omega_m^{(2)}(F)$$

$$\omega_k^{(1)}(F) = \frac{3}{2}nF \qquad \omega_k^{(2)}(F) = \frac{3}{8}kn^4F^2 \qquad \omega_m^{(2)} = \frac{9}{8}mn^4F^2$$

- $\omega_{\pm}$ depend on *n* and F - to first order precession reverses when reverse F
- second-order terms prevent perfect rephasing - minimize using low-m,k states
- consider behavior in rotating frame

Evolution of Pseudo-Spin Probability Density Distribution

- shown in rotating frame
- consider x,y components J_+'
- distribution broadens due to dephasing
- pronounced rephasing (echo) following field reversal
- quantify dephasing by considering excess width in azimuthal angle

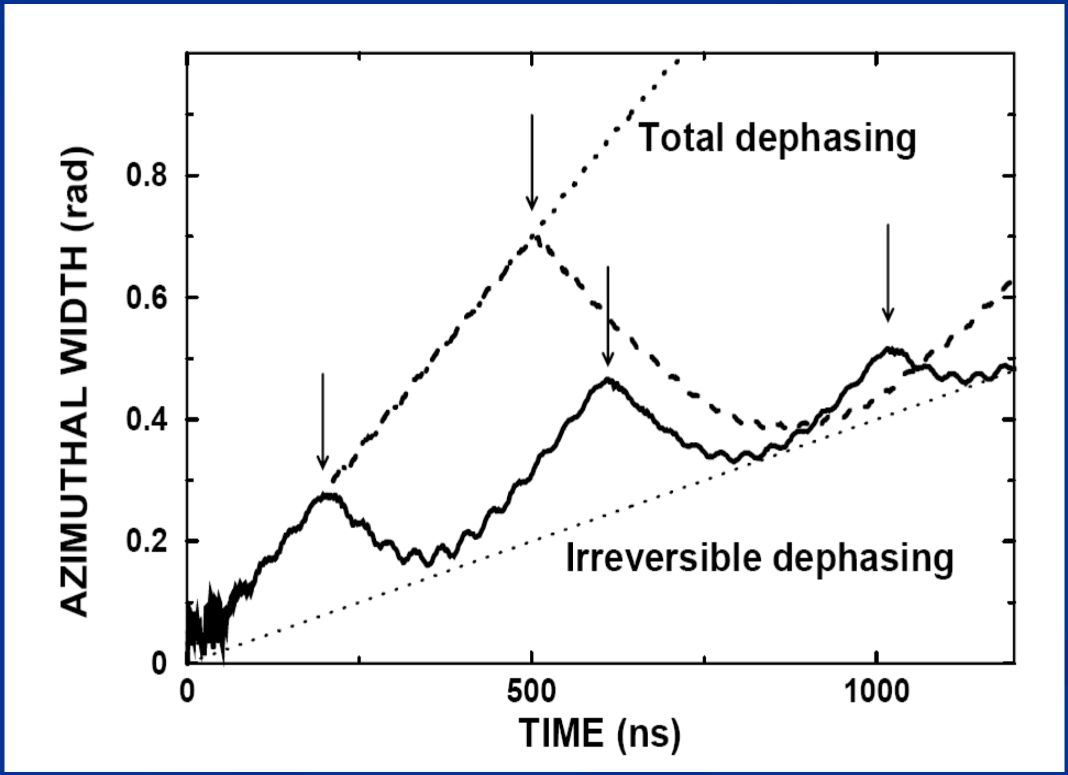


(a) superposition of extreme parabolic states k = n-1, 342 < n < 358. (b) initial experimental state

Characterization of Dephasing

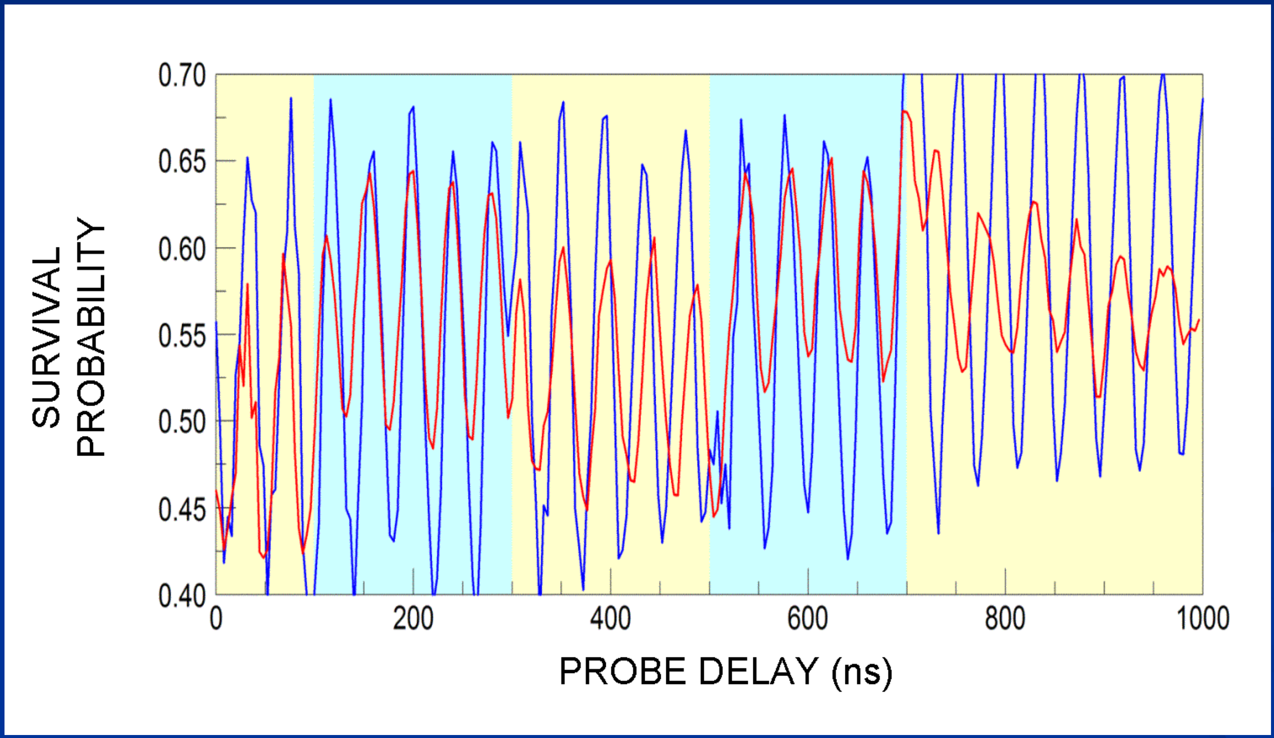
Quantify through increases in azimuthal width

- azimuthal width grows linearly in time
- dephasing associated with second-order terms irreversible
- can limit dephasing using periodic reversals



Effect of Periodic Reversals

- field reversed at 100, 300, 500, and 700 ns

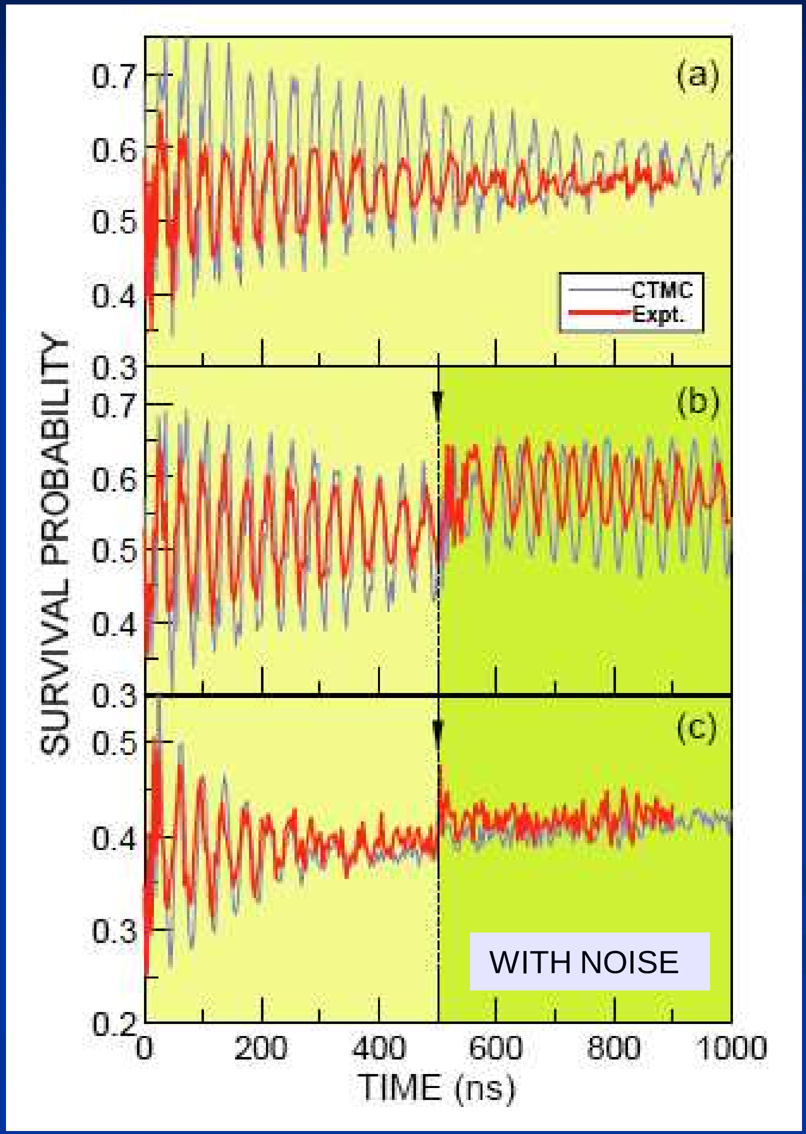


- strong quantum beats seen even at late times
- reduced amplitude provides evidence of irreversible dephasing

Noise-Induced Irreversible Dephasing: Decoherence

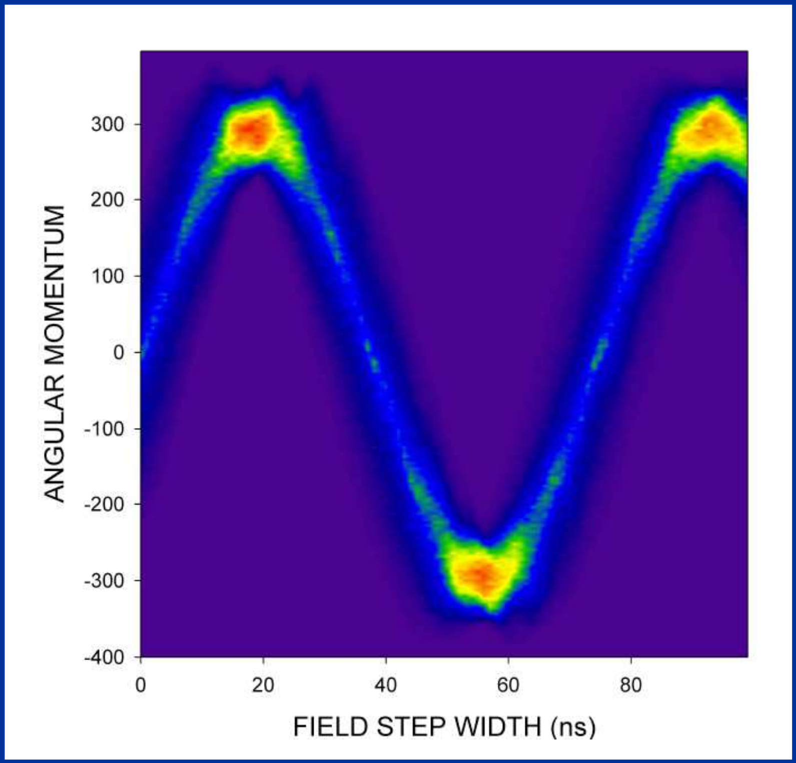
- colored noise produced by pseudo-random pulse generator
- presence of ±10% amplitude noise damps quantum beats and destroys the echo - introduces irreversible dephasing - decoherence
- can examine effect of the noise frequency spectrum

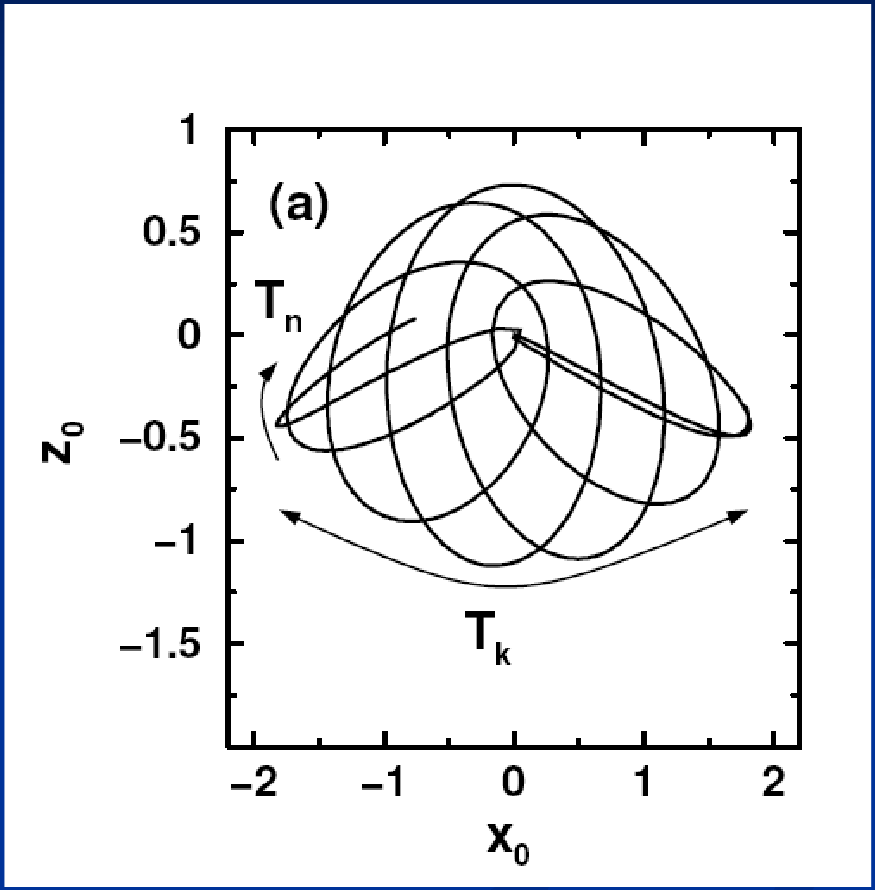
Stark echoes allow exploration of decoherence in mesoscopic systems on timescales shorter than revivals



Production of quasi-2D near-circular states

- create quasi-1D n = 350 state oriented along x axis
- apply dc field step in z direction
- turn off when L maximum

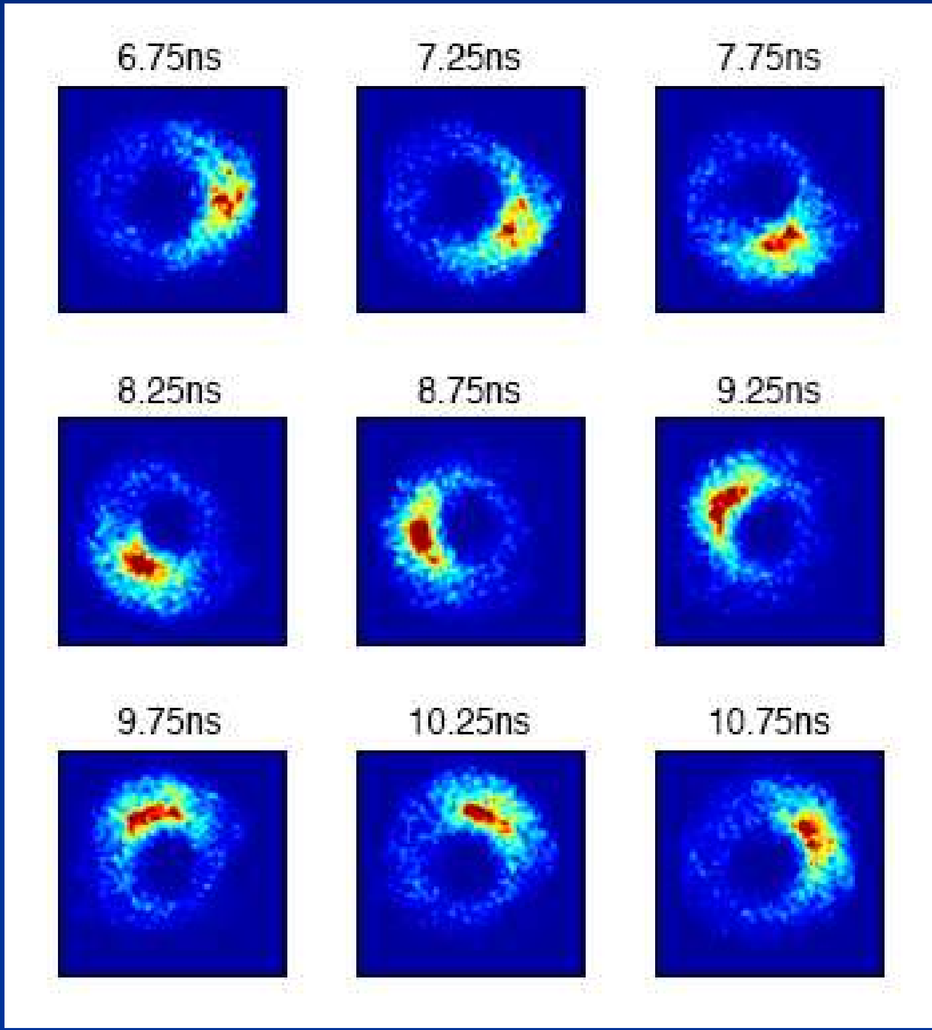




- produce localized wavepacket in near circular “Bohr-like” orbit

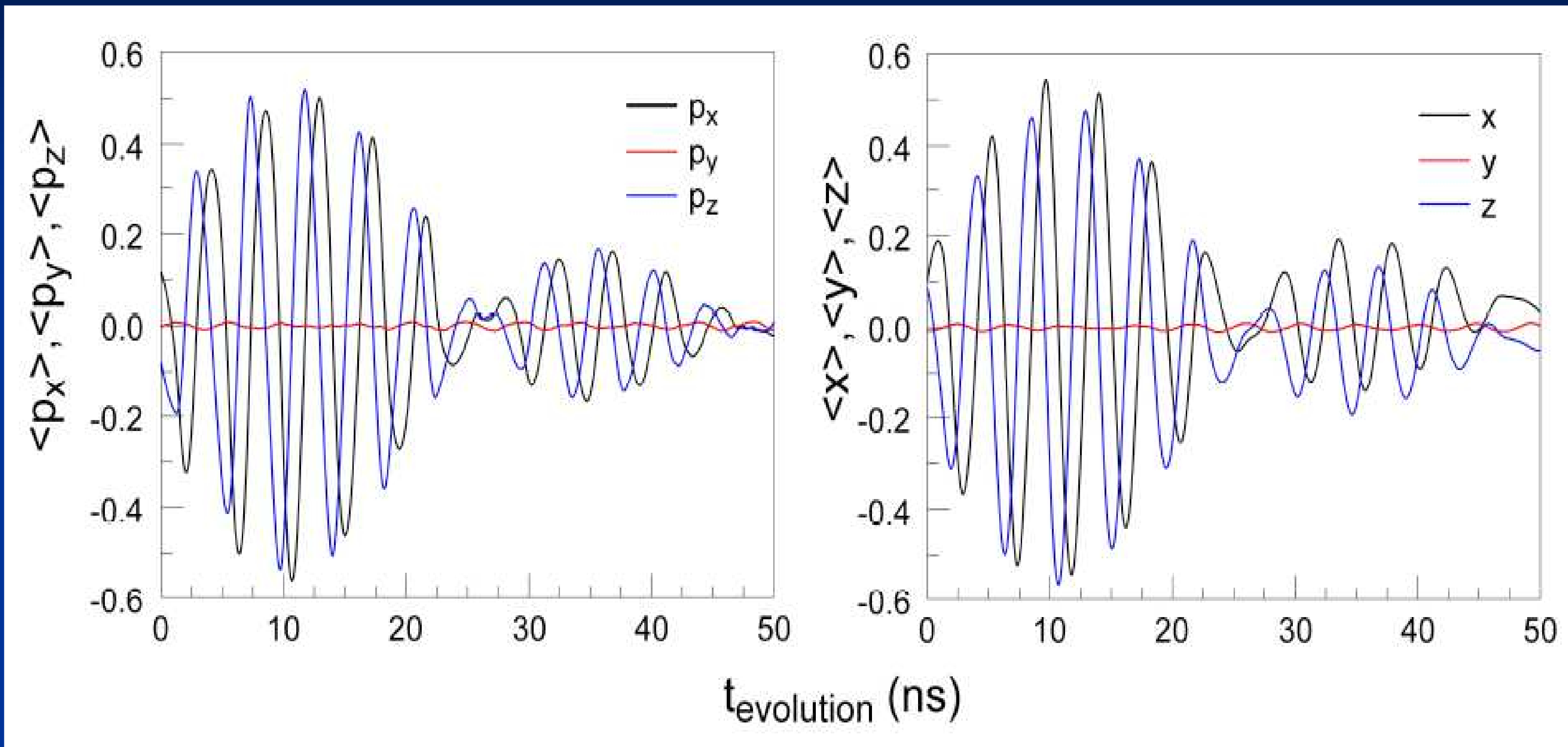
Wavepacket evolution

Apply dc field of 20 mV cm^{-1} to quasi-1D n = 306 atoms for 22 ns - follow subsequent behavior using CTMC simulations



- wavepacket remains localized as “orbits” in xz plane
- mimics the original Bohr model of atom
- follow evolution through behavior of <x> , <y>, <z> and $<p_x>$, $<p_y>$, and $<p_z>$

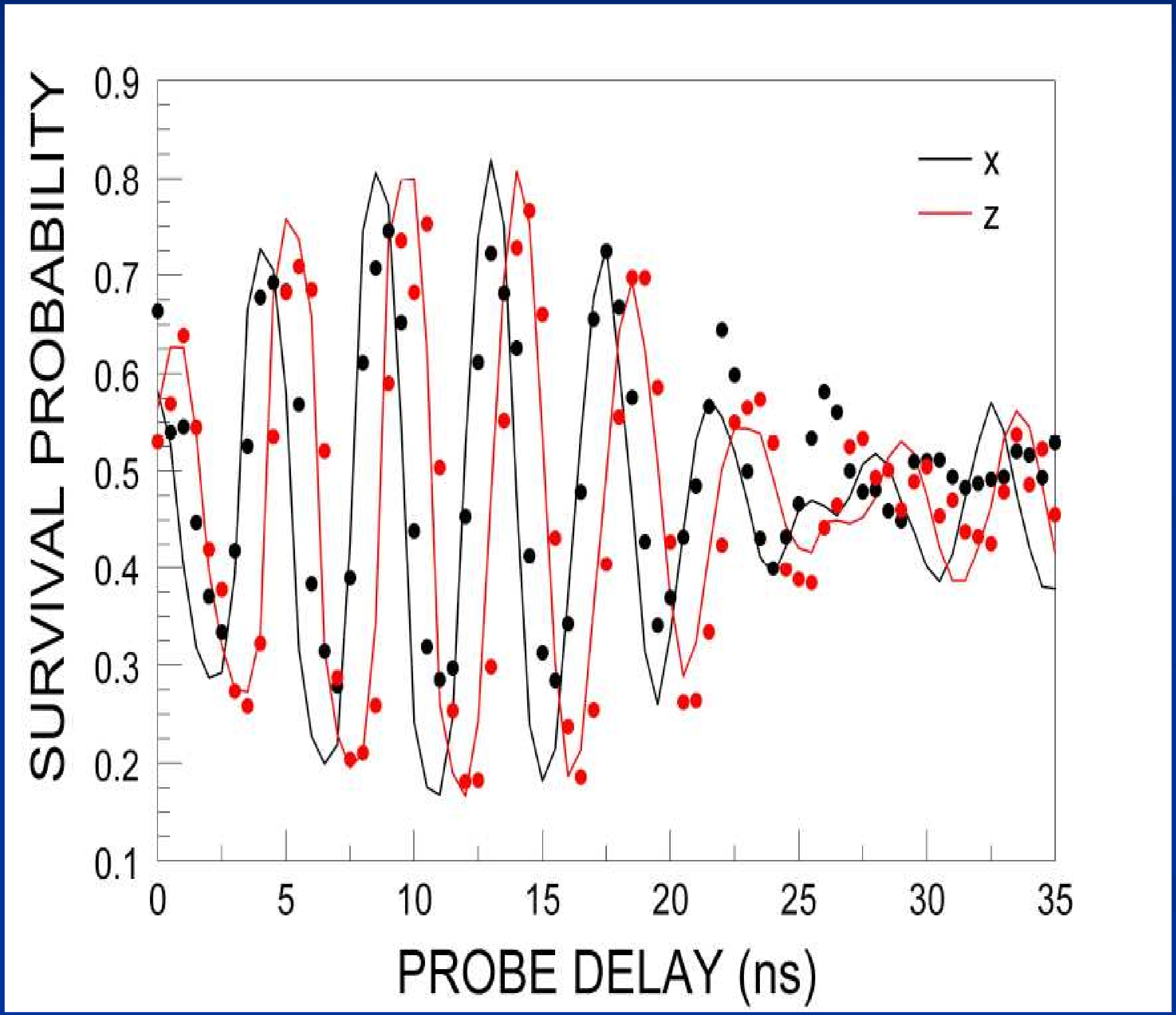
Wavepacket evolution: simulations



- strong variations in $\langle p_x \rangle$, $\langle p_z \rangle$ - 90^0 out of phase
- $\langle p_y \rangle$ ~ constant - motion in xz plane
- strong variations in $\langle x \rangle$, $\langle y \rangle$, and $\langle z \rangle$ - 90^0 out of phase
- produce near-circular states

Circular atoms - experiment

n = 306, 20 mV cm^{-1} field applied for 22 ns



- strong oscillations 90^0 out of phase
- good agreement with simulations
- produce near circular “Bohr-like” states
- enables range of new dynamical studies

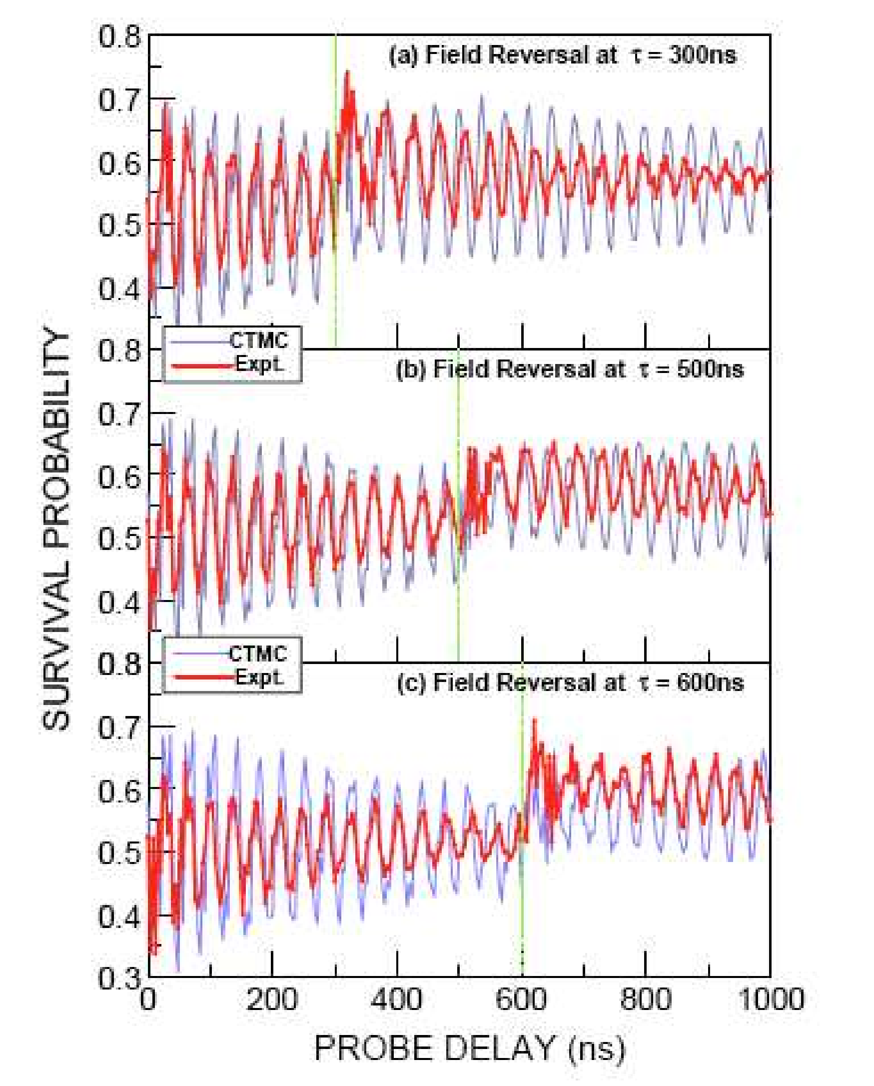
Conclusions

- can control and manipulate Rydberg wavepackets with remarkable precision using HCP trains
- Stark quantum beat echoes provide sensitive probe of reversible and irreversible dephasing
- Rydberg atoms form a valuable bridge between the quantum and classical worlds

Electric Dipole Echoes: Effect of Reversal Time

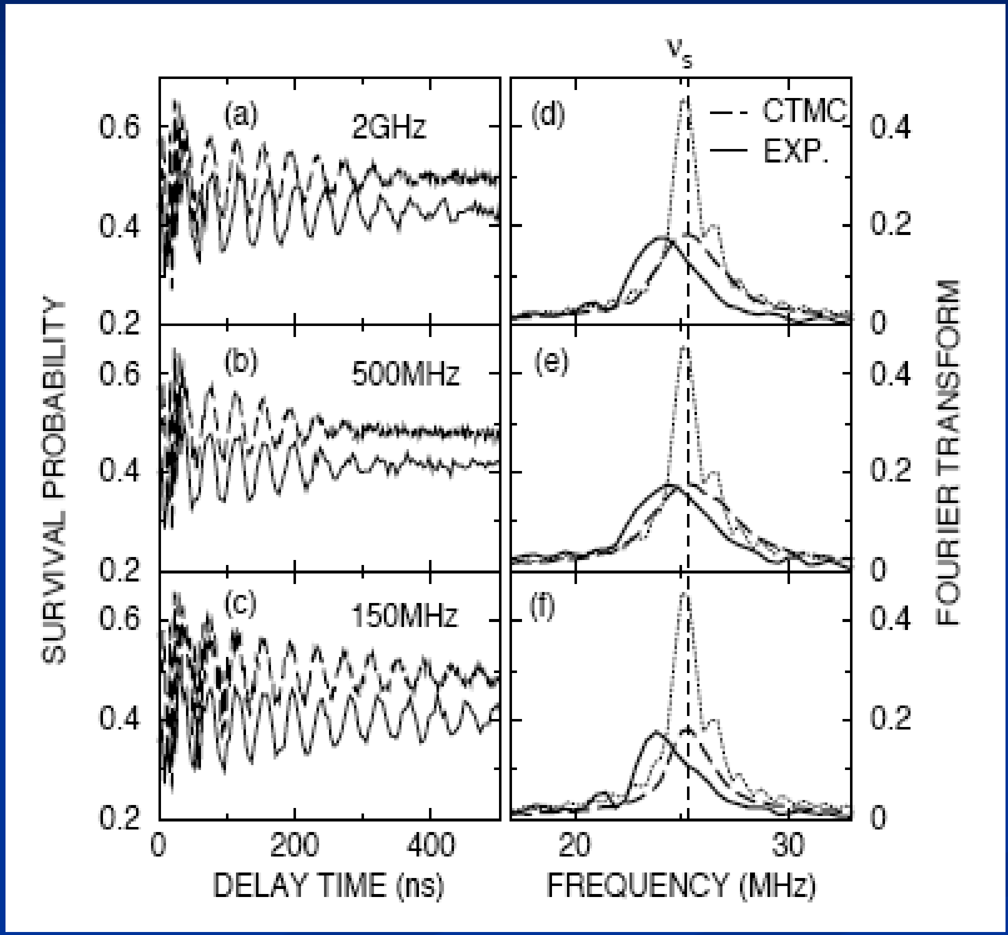
- strong quantum beat echo at t ~ 2τ
- echo shows initial dephasing reversible and largely coherent

Origin of effect ?



Stark wavepackets: effect of noise “frequency”

- damping depends on time bin width T_{ran} and related characteristic frequency $\nu_{ran} = 1/T_{ran}$
- evident from width of Fourier transform
- decoherence greatest when ν_{ran} ~ twice orbital frequency

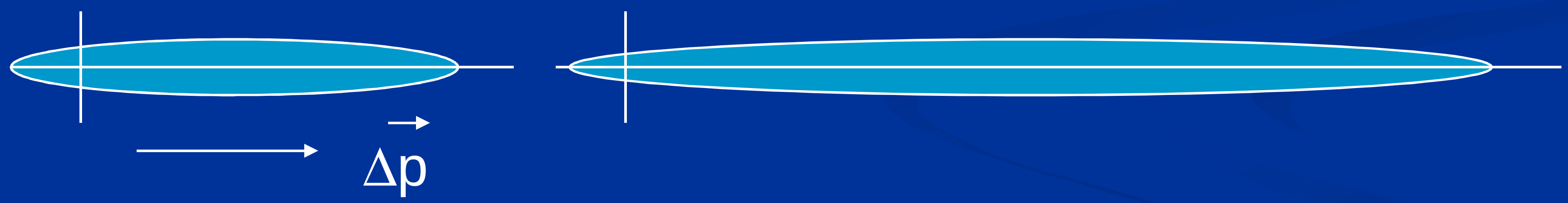


Also explore decoherence through Stark quantum beat echoes

Atomic engineering

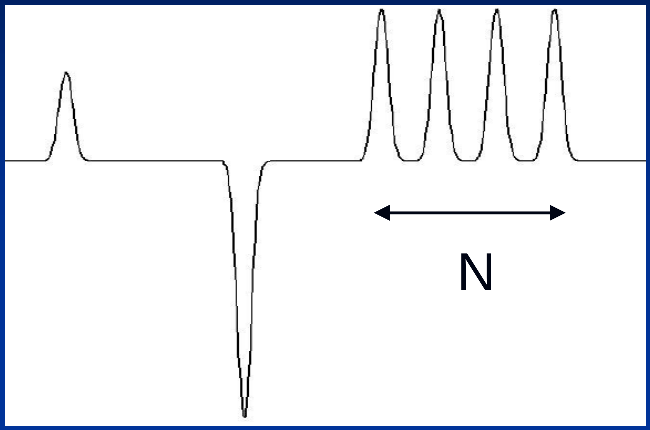
Use phase-space localized state and tailored HCP sequence to engineer targeted final states - very-high-*n* (*n*~600) quasi-1D atoms

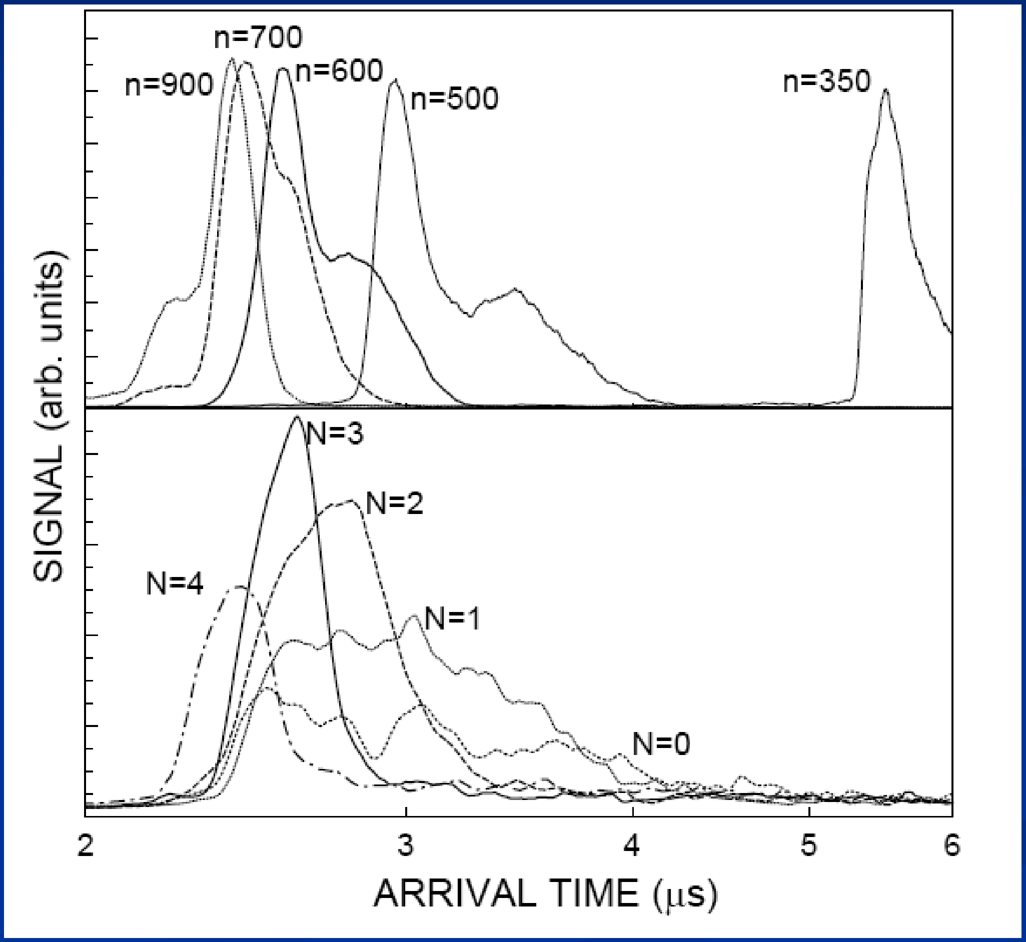
Apply strong kick in +z direction to localized quasi-1D *n*~350 atom



Even with pre-localization populate broad distribution of final states - paradoxically can narrow by application of further HCPs

Production of quasi-1D very-high-*n* states

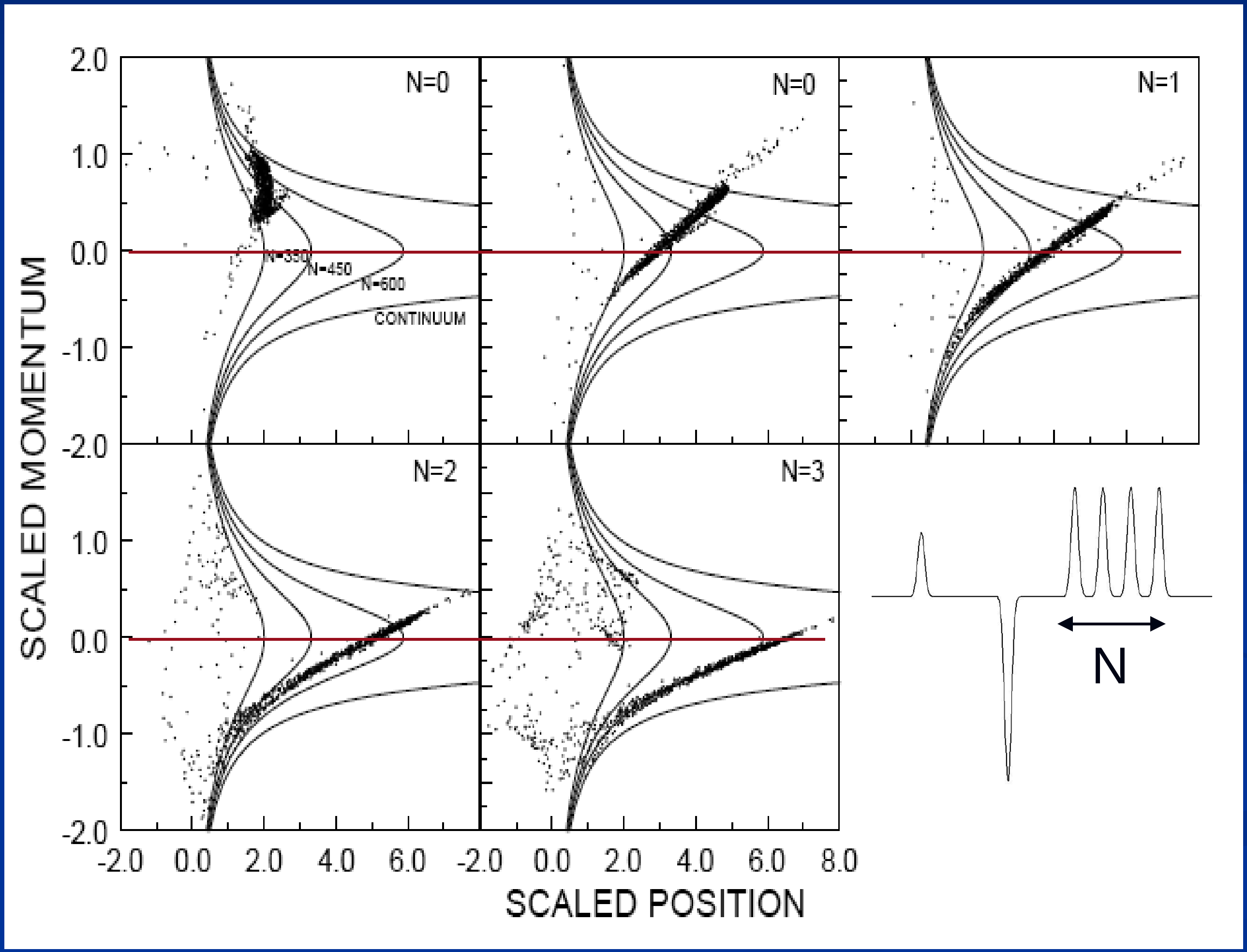




- use SFI to measure final *n* distribution
- observe initial narrowing of *n* distribution as N increases - counter-intuitive!
- confirmed by CTMC simulations - demonstrate origin of effect

Physical origin of *n* focusing

Phase-space portraits describing evolution of 1000 initial trajectories



- strong n focusing after N~3 kicks
- *n* distributions controlled with HCPs
- improve control with genetic algorithms

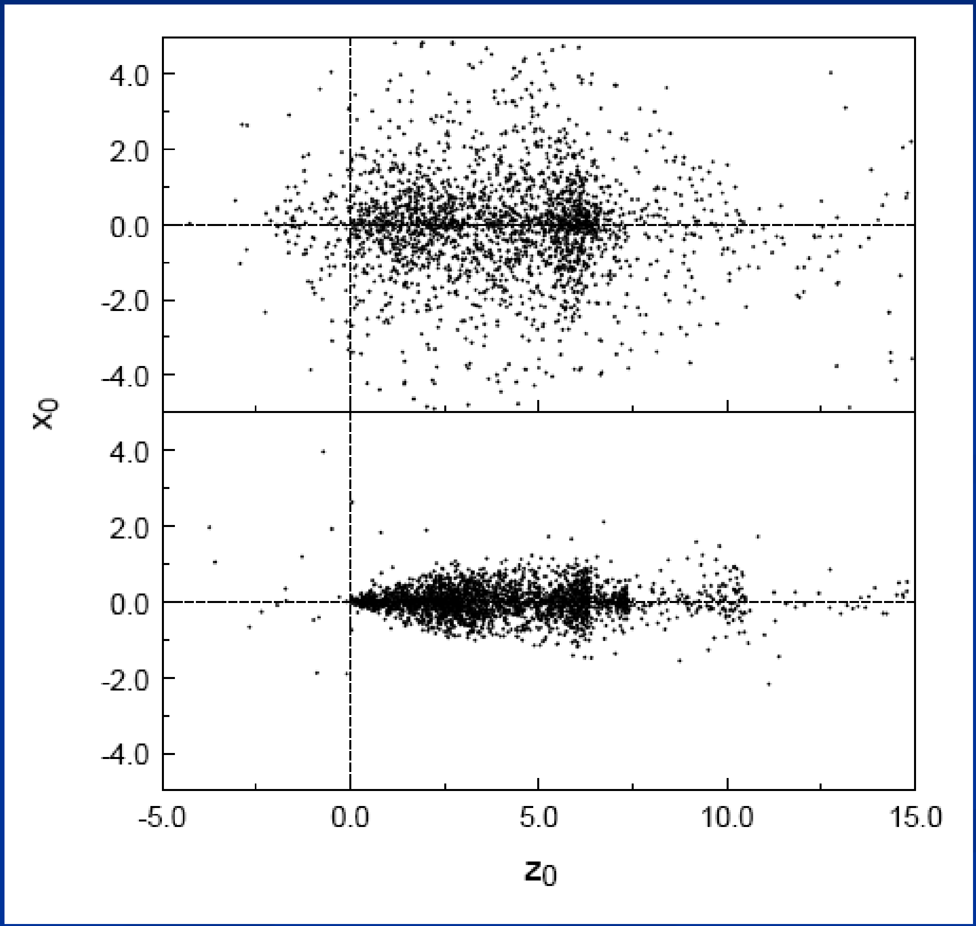
Product states: spatial distribution

N=3 HCPs, 120ns delay

Produce quasi-1D very-high-*n* atoms

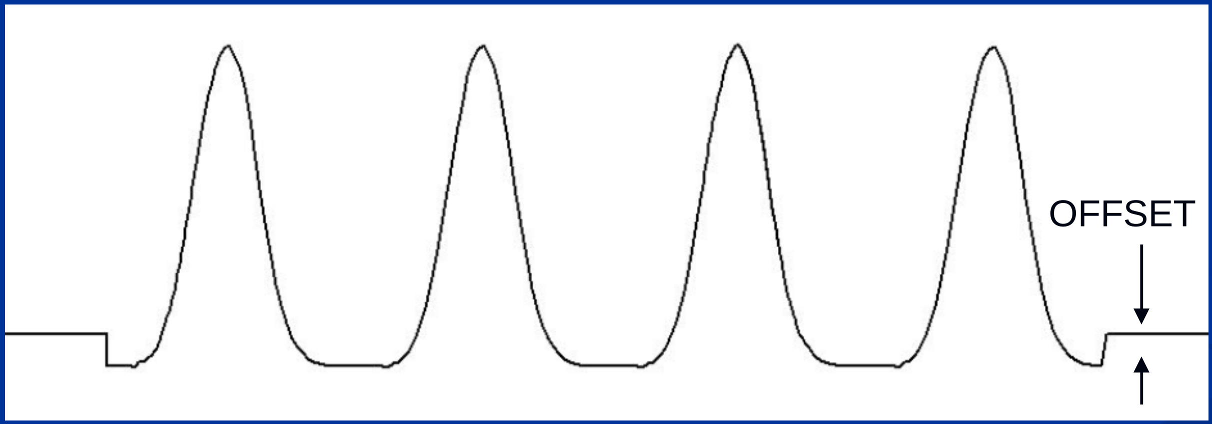
Enable studies at high scaled frequencies ν_0~15 where:

- observe novel behavior in survival probability
- see effects of quantum localization



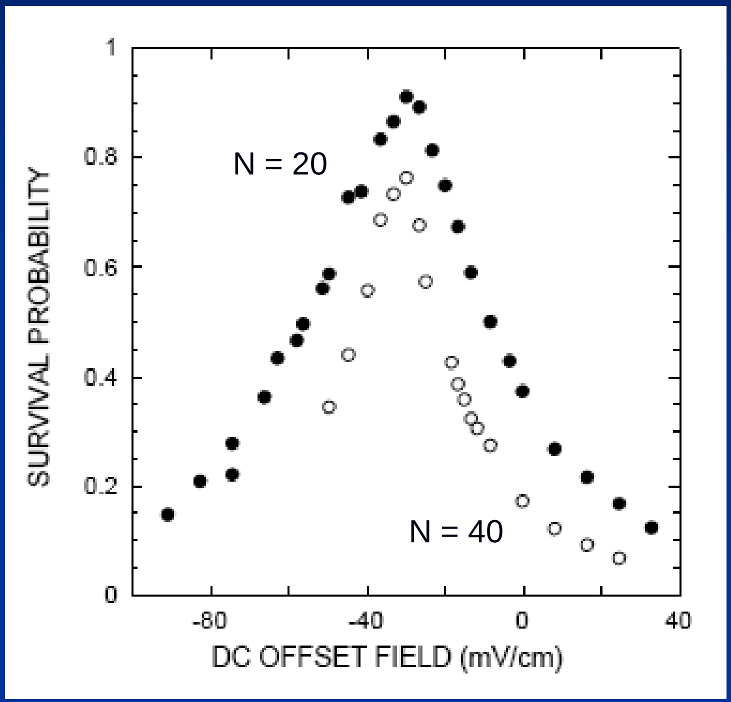
Control of low-lying states

- use femtosecond lasers and high-harmonic generation to produce trains of attosecond HCPs
- freely propagating - no net dc field present
- investigate effect by applying offset bias during HCP train

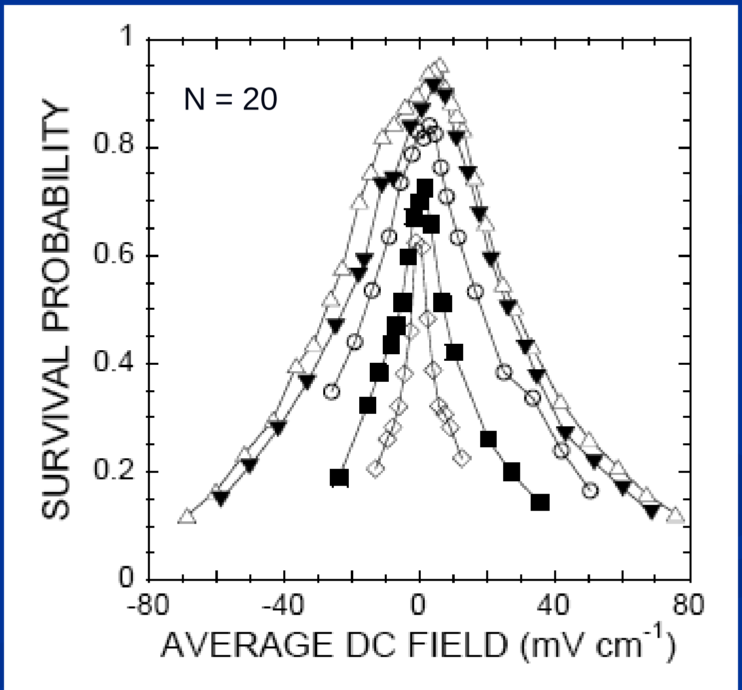


- offset field dramatically changes atomic response to HCP train

Effect of offset field: K(350p)



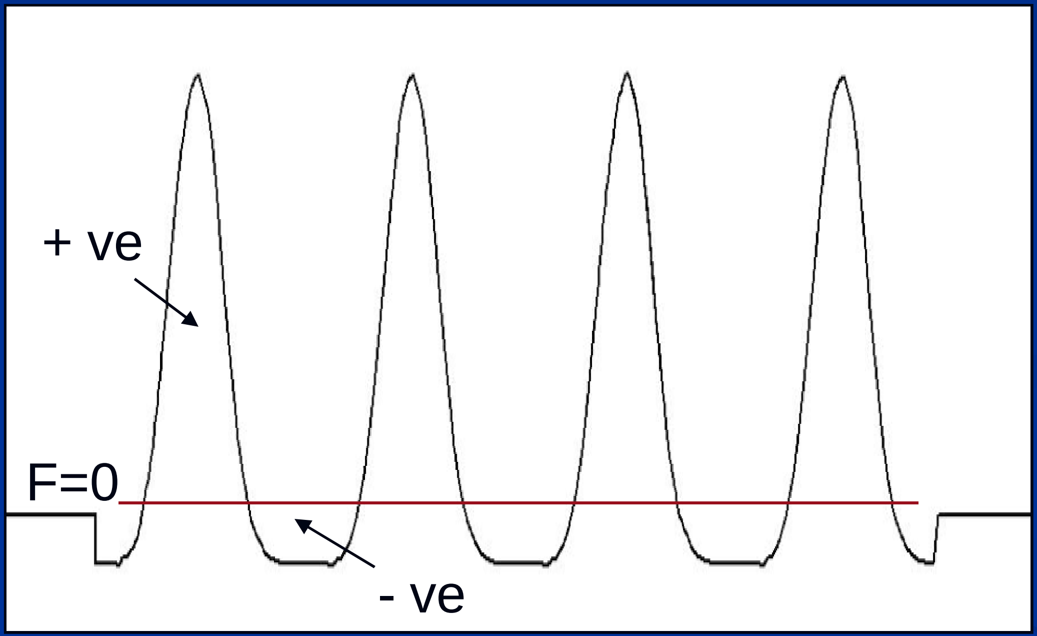
- survival probability depends on offset field



- for ν_0 ~ 0.3 - 3.3, survival probability maximum when F_{av} = 0

Origin of peak in survival probability at $F_{av} = 0$

- expected for $\nu_o >> 1$ - positive and negative kicks cancel



- picture less clear for $\nu_0 << 1$ but origins similar

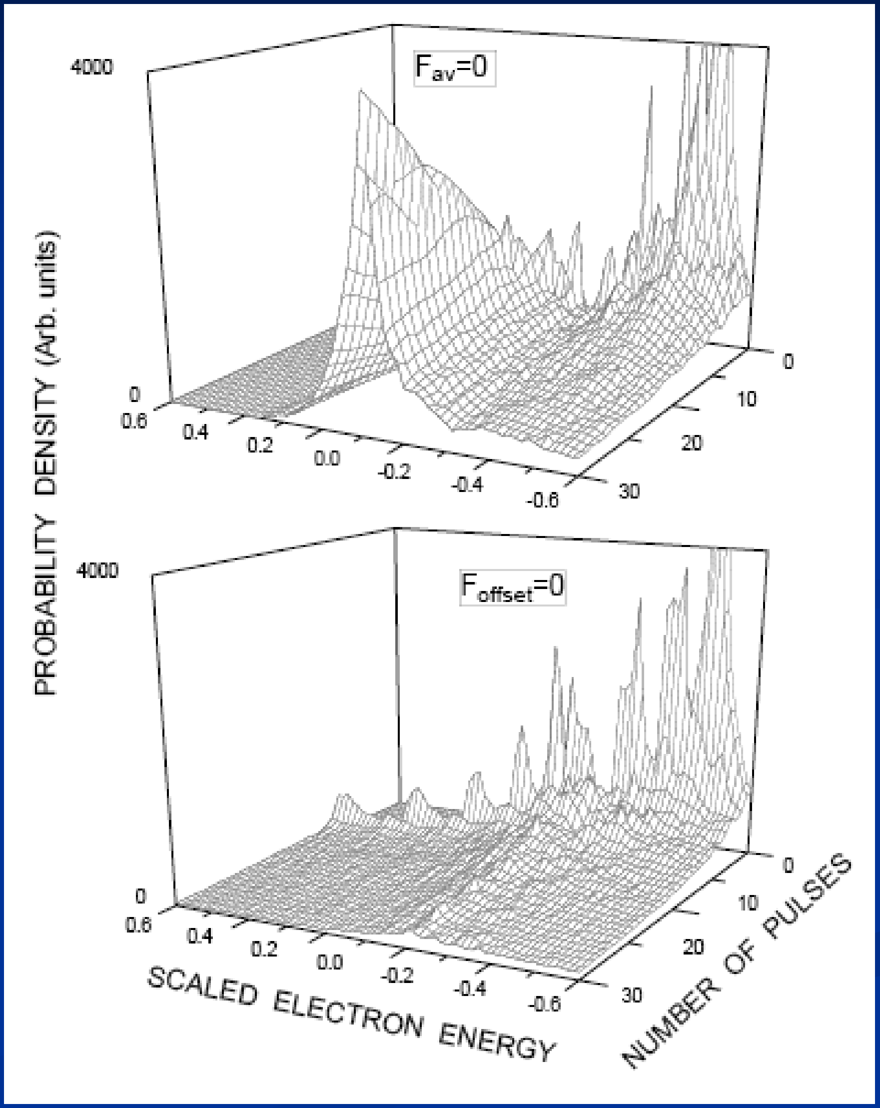
Evolution of electron energy distribution:

K(350p) ν_0=0.25

Observe:

- population trapping near continuum
- effects of dynamical stabilization masked
- dynamics strongly influenced by presence of offset field

Similar behavior seen with bidirectional kicks

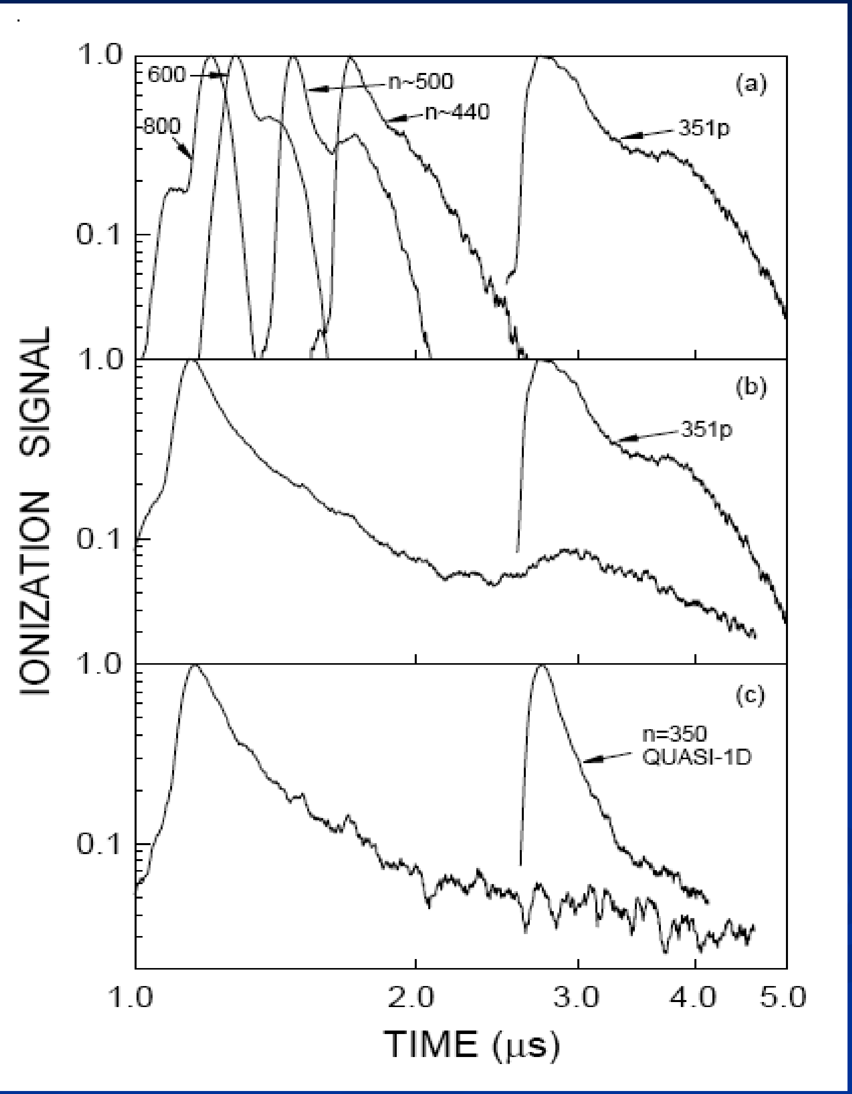


Bidirectional kicks: SFI profiles

Average field experienced
by atoms is zero

- observe population trapping near continuum for 351p and quasi-1D states
- peak near parent *n* for 351p state due to trapping in quasi-stable island for ν_0~1

See evidence of this trapping in CTMC simulations



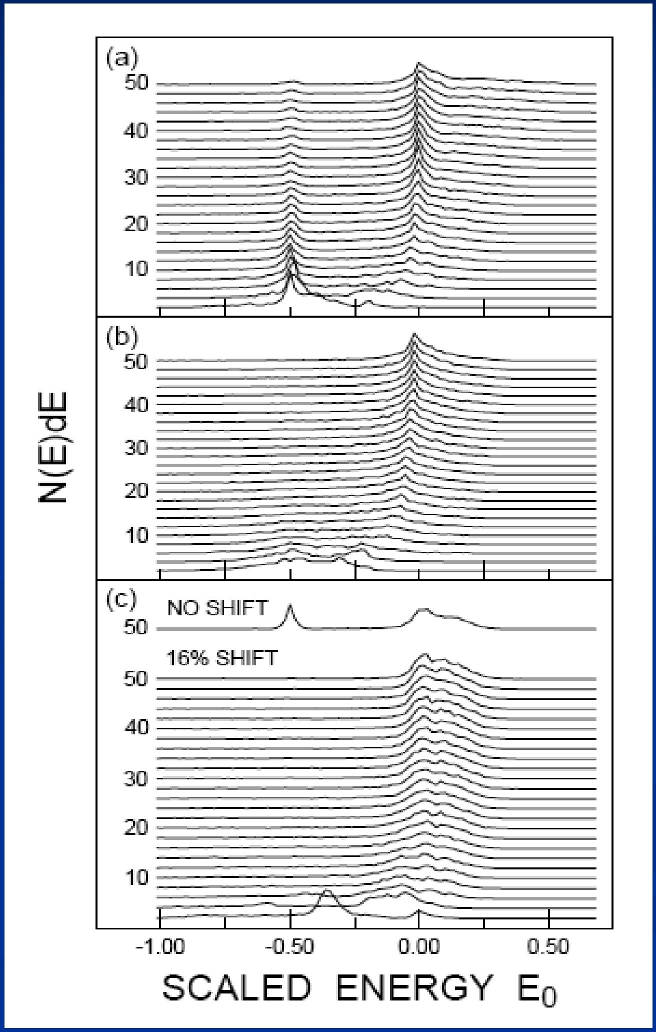
Bidirectional kicks: CTMC simulations

- population builds up near continuum
- for ν_o~1 feature persists near parent state energy - trapping in quasi-stable island

350p
$\nu_0 = 1$

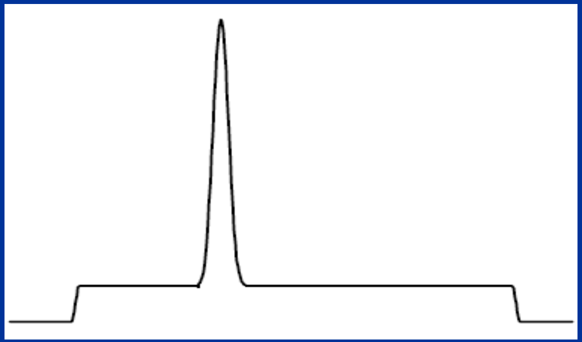
350p
$\nu_0 = 3$

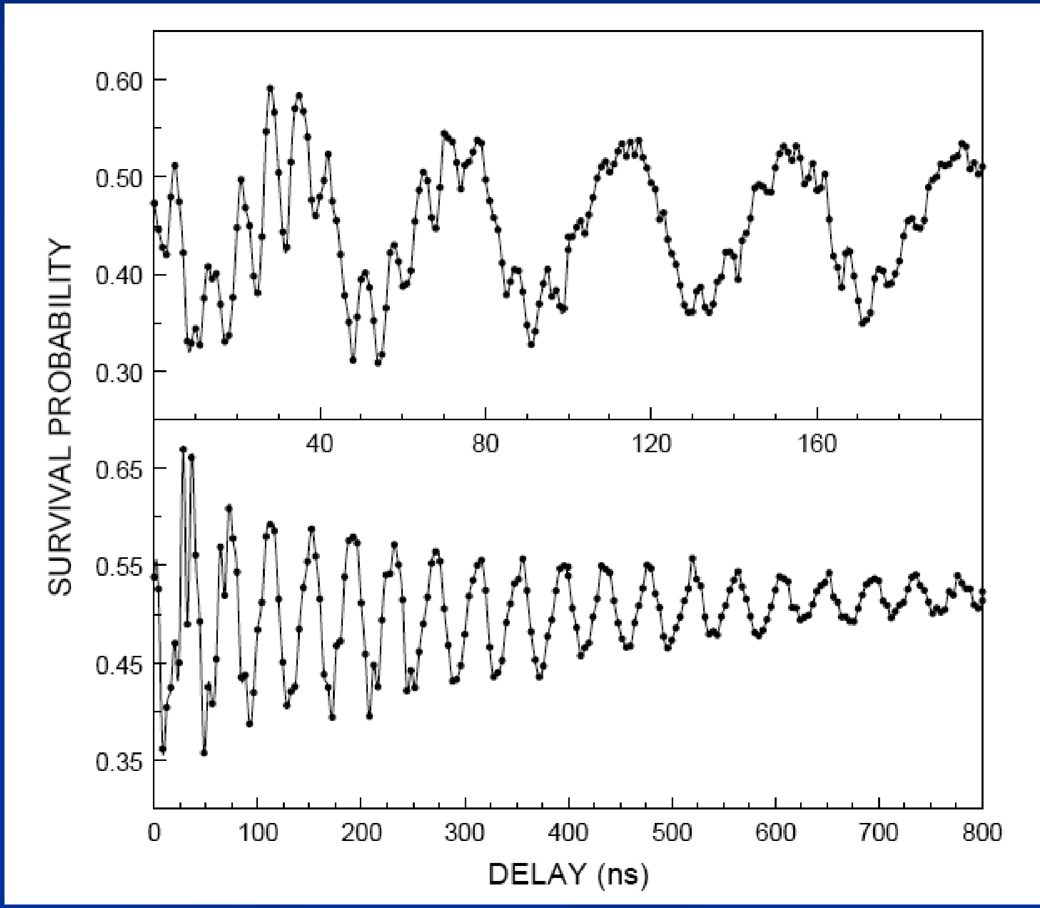
Q-1D
$\nu_0 = 1$



Dephasing of Stark wavepackets

- create by applying field step to K(350p) atoms
- monitor evolution with delayed probe HCP





- observe dephasing

Stark wavepackets: noise-induced dephasing

Noise source: generator delivering random sequence of 0s and 1s at frequencies up to 3GHz, amplitude 10% of field step

Observe:

- strong noise- induced damping of quantum beats
- damping rate depends on noise “frequency”
- results well reproduced by simulations

Explore nature of dephasing by looking at quantum beat “echoes”